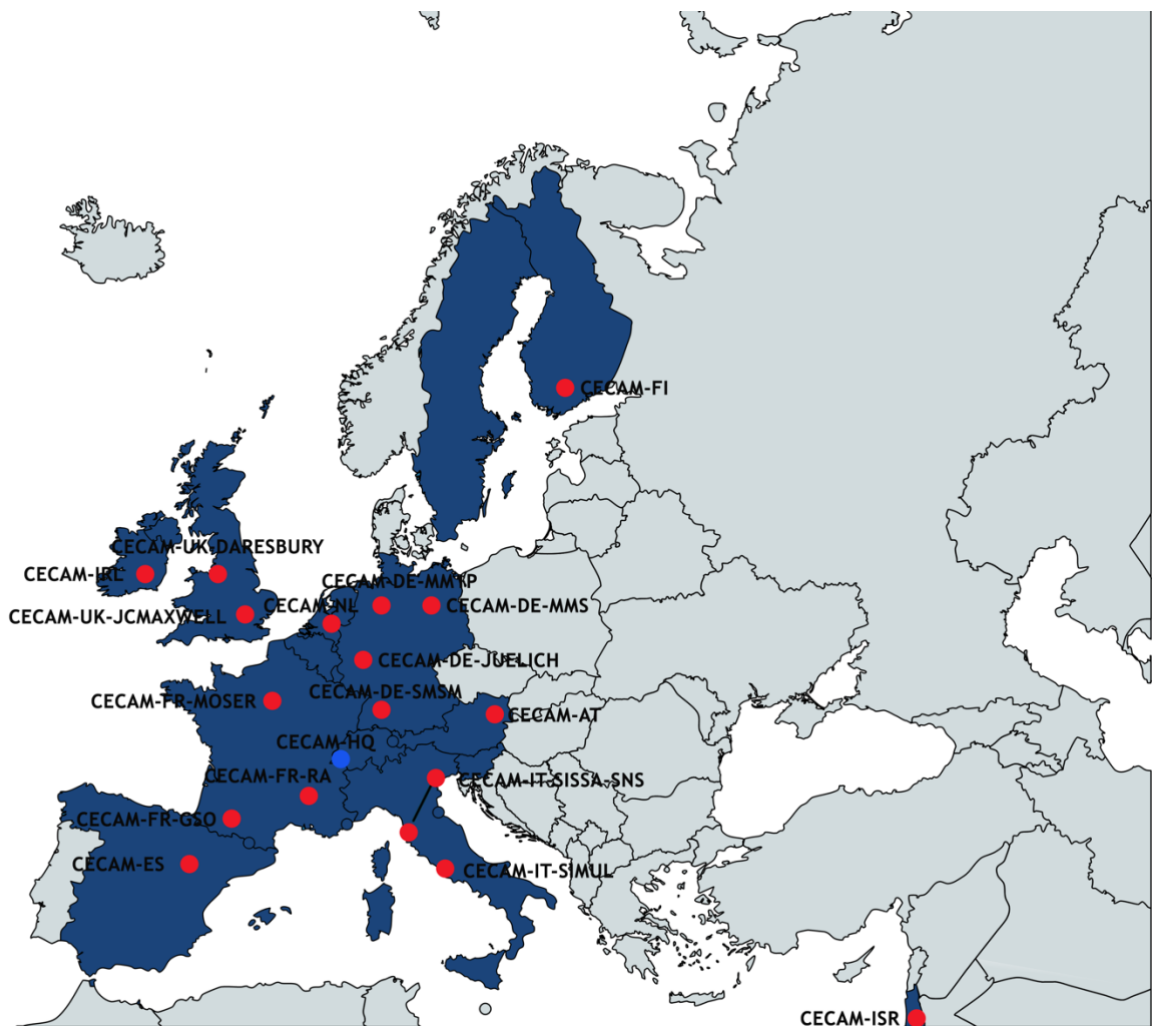


# Scientific Report

# 2019



2019 was a special year for CECAM, marking the fiftieth anniversary of its foundation. Our rich and diverse program of activities is perhaps the best testament to the vision of Carl Moser and the early animators of our growing community.

The 2019 flagship program brought 2561 participants (22% women) to the 55 workshops and 20 schools hosted by the 18 CECAM nodes and the Headquarters (HQ) at EPF-Lausanne. In particular, HQ hosted 15 workshops, one of which in the Swiss partner Università della Svizzera Italiana (USI) in Lugano, and 4 schools – two of which in USI. 40 workshops and 16 schools were hosted in the nodes. In addition, 5 Extended Software Development Workshops, 2 State-of-the-art workshops and 1 Scoping Workshop for industrial outreach were hosted across the network in the framework of the [E-CAM project](#), a Center Of Excellence for Computing Applications funded by the European Commission, and coordinated by CECAM. The reports describing the main scientific output of the programme are collected in this document.

The fiftieth birthday of CECAM was celebrated with the [dedicated conference](#): “Molecular and materials simulation at the turn of the decade: Celebrating 50 years of CECAM” that took place in Lausanne on September 9-12 2019. 8 plenary and 4 daily parallel sessions, with 48 invited and 48 contributed presentations, animated the event that gathered more than 300 participants. The conference offered also the ideal venue for the remise ceremony of the 2019 Berni J. Alder CECAM prize, awarded to Prof. Sauro Succi for his seminal contributions to the Lattice-Boltzmann method. The conference programme, book of abstract and list of participants are also included in this scientific report and illustrate the broad set of topics that are now an integral part of CECAM scientific domains, together with the scientific excellence of its community. Finally, the first chapter of CECAM’s history was reconstructed by Giovanni Battimelli in “A European Research Center in Orsay: CECAM 1969-1993”. This publication is also enclosed in this report.

We are, as always, grateful to the organizers and participants of our workshops and schools for continuing to bring to CECAM exciting science and foster advancement through discussion, training, and an open and enthusiastic approach to new challenges in simulation and modelling. With their help, and with the precious assistance of the many leading scientists in our broad community that assume roles of responsibility in the network, we shall continue to provide useful and high-quality service to scientists in our domain with unchanged commitment.

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# Modeling tribology: friction and fracture across scales

Location: CECAM-HQ-EPFL, Lausanne, Switzerland

Webpage: <https://www.cecarn.org/workshop-details/199>

Dates: Jan 28, 2019 - Jan 30, 2019

## 1 State of the art

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Tribology, the science of contacting surfaces, remains poorly understood. The challenge lies in the multi-scale and multi-physics nature of friction. Tribological systems may cover: (i) Length scales from nanometers (atomistic and molecular mechanisms at micro- or nano-contact levels) to kilometers (fault seismicity) (ii) Time scales from picoseconds (tribo-induced chemical reactions) to years (earthquake cycle) (iii) A variety of coupled processes including phase changes, fluid flow, complex rheologies of the contacting materials, fragmentation and third body formation. Our ambition has been to clarify the nature, validity range and coupling of the many elementary processes involved in tribological systems at all space and time scales. A special emphasis has been put on identifying how the tools used in the fracture mechanics community (e.g. equation of motion for crack propagation, size-dependent fracture modes, rupture of heterogeneous media) can help in describing tribological processes. In this context, about 40 researchers from the Physics, Geophysics, Mechanics and Engineering communities have joined the workshop and discussed the following key topics:

1. Onset of sliding: static friction, micro-slip fronts, analogy with fracture mechanics, role of interfacial heterogeneities at all scales
2. Geologically-relevant friction processes: modeling friction-related processes in earthquakes, avalanches or glaciers, rock failure
3. Elementary wear mechanisms: erosion, debris formation, atom-by-atom material removal
4. Dissipative processes at interfaces: adhesion, viscoelasticity, plasticity, radiations, and their coupling in tribological systems
5. Transient processes: instabilities, impacts, unsteady kinematics, short time scales phenomena
6. Statistical aspects: fluctuations beyond the most likely behavior, predictability, intermittency, size-dependence and scaling laws

## 2 Major outcomes

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The workshop was organized in six sessions, in decreasing order of scales. Friction processes at geological scales: The discussion started with recent laboratory experiments, which highlighted outstanding open challenges. Amongst which we highlight: How common is dynamic weakening on natural faults? Do high-velocity laboratory experiments promote dynamic weakening (through enhanced shear localization or inefficient heat transport)? Can laboratory experiments incorporate effects such as geometric complexity of single faults, off-fault dissipation and fluid pore pressure evolution, networks of faults? More generally, do scale-specific effects limit our ability to extrapolate lab results to geological scales? Onset of sliding: In stick-slip regime, the onset of sliding is mediated by slip fronts, the propagation speed and length of which are well described as mode II cracks. However, the conditions at which different

kinds of fronts can arise are still debated. The question of front nucleation appears as one of the outstanding open question. There are currently two main descriptions of interfaces at the onset of sliding, either as a continuum medium, or as a set of discrete, disordered micro-contacts. Coming with a combined, multiscale description is a clear challenge. Statistical/non-linear aspects: insights from fracture Linear Elastic Fracture Mechanics (LEFM) has demonstrated its power in addressing the growth of single brittle cracks, in both tensile (mode I) and shear (mode II) loading configuration. Yet the physics of ductile fracture is less advanced, as this involves more material parameters. Progress is expected in the coming years. Opportunities exist in understanding crack branching instabilities. Recent developments in experimental and numerical techniques have also been discussed. X-ray micro-tomography appears as a unique probe to image in 3D at small scales damage and fracture processes in a wide range of materials. In numerical modelling, although phase field approaches are popular, they should be used with care in friction, where dissipation is not localized. Mechanics at rough contacts: Rough contacts should be understood as an intermediate scale (mesoscale). An opinion shared by many is that this scale holds great promise to relate dissipative mechanisms at micro-contacts to macroscopic friction laws. Elastic normal contact of rough surfaces is now reasonably well understood. But a major challenge is to understand how these micro-contacts change in the presence of sliding, roughness evolution, and plasticity. Elementary processes & coarse-graining: Two major points have been pointed out: (i) The need of model experiments at the mesoscale (between continuum models and discrete descriptions) to understand the underlying mechanisms and validate numerical models; (ii) The need for technologically useless but simple materials. The difficulty to get funding along the above lines has been raised. The possible outcomes and difficulties of machine learning and data mining techniques have been discussed. Tribology at small scales: In molecular dynamics, the discussion emphasized the need to work not only on more advanced, system-specific potentials, but also on simple but versatile potentials used to study generic elementary processes. The community still faces difficulties to find (model) experiments amenable to direct comparison with nanoscale modelling, partly due to simulations being limited to very small times scales. Big data: Although not explicitly part of the programmed discussions, the challenges raised by big data in science and machine learning have been repeatedly mentioned: either as an unavoidable difficulty due to the increasing amount of data produced by high resolution imaging (fast cameras, tomography) and the necessity to use/develop tools to analyze and store those data ; or as an opportunity to guide scientists' intuition by exposing otherwise hidden or unexpected correlations.

## 3 Community needs

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At several instances during the workshop, we discussed the convergence of model (discovery) experiments and model simulations. To accelerate this convergence there is a need for open science/open data platforms. Local initiatives are already taking place such as release of open-source software and European networks for sharing experimental database. CECAM could take a leading role in centralizing such efforts. This workshop was unique in facilitating networking in tribology. We have brought together a diverse group of leading scientists in geophysics, physics, materials science, and engineering, from geological to atomic scales. Also unique to our workshop was that each session started with a keynote from an experimentalist, whose role was to highlight specific experimental advances motivating a need for cross-fertilization with numerical modeling and theory. This resulted in intense (sometimes heated, but friendly) discussions, with consequences and collaborations that will extend far beyond this workshop. The community would welcome a CECAM series on this topic. The societal and economic impact of tribology as well as emerging scientific questions necessitate a dedicated forum within CECAM, much like the dedicated Tribology Gordon conference in the

US. We wish to highlight that we had to decline the participation of many people (about 30), several of them were from leading industrial groups. The workshop was strictly invitation-only, and in fact all the persons we contacted -but one- agreed to come. This shows that the community is in great need of such a forum.

## 4 Funding

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The difficulty has been raised to fund model (real or numerical) experiments on simple and well-controlled systems (hence far from technological applications). These analog experiments constitute an obligatory step to isolate the key mechanisms at play in tribology problems, and subsequently control/limit their effects in the real situations of technological interests. While not explicitly addressed during the workshop, the setting of an ITN network on this thematic could be timely: The participants and in particular the PhD students attending the workshop found it very useful to discuss and confront the problems with the different communities represented at the workshop (seismologists, physicists, engineers, and materials scientists).

## 5 Will these developments bring societal benefits?

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Today, about 25% of the energy need worldwide is actually used in tribological contacts, either as friction losses or to remanufacture worn pieces of equipment (K. Holmberg & A. Erdemir, *Friction* 5(3): 263–284, 2017). The potential of new materials, surfaces and lubricants, including those discussed during the workshop, is huge, as they could enable about 40% reduction of those losses within 2035. Such a reduction would obviously reduce the corresponding greenhouse gases emissions, thus offering both economical and societal benefits. Health wise, the control of wear is also a major challenge, as the various wear processes, especially in the transportation industry, emit a large amount of fine particles in the environment. For instance, tires, roads and brake pads abrasion, amount to about 40% of the polluting particles in major cities. The advances in the understanding of wear discussed during the workshop open the way to reducing those particle productions, and hence approaching further EU's ambitious reduction goals. Finally, the advances on the prediction of interfacial failure discussed during the workshop will contribute to the advancement of a better predictability of natural disasters, like earthquakes, landslides and avalanches, in turn leading to a reduction of the associated human casualties.

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# Multiscale modeling from macromolecules to cell: opportunities and challenges of biomolecular simulations

Location: CECAM-HQ-EPFL, Lausanne, Switzerland

Webpage: <https://www.cecarn.org/workshop-details/241>

Dates: Feb 04, 2019 - Feb 06, 2019

## 1 State of the art

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The field of molecular simulations is experiencing a transformative phase, due to ever increasing advances in structural biology. Because of these, we are currently witnessing an explosion in the number of large and complex biomolecular structures deposited into the protein databank. These are solved at increasing resolution, but often only provide a static picture of the system, which call for further interpretation by computational methods. At the same time, there are still plenty of cases where a single experimental approach cannot lead to full structural description of a system. In such cases, data from various sources must be combined with computational models of various complexities and scales in order to shed light on biological function. The goal of this workshop has been to discuss the current challenges of multiscale simulations, and the need of going beyond the limits of time scale and system-size, in order to model biological systems from the atomistic to the cellular level. Molecular simulations from atomistic to coarse grained (CG) and mesoscale (MS) representations of biomolecules into the cell environment have been discussed, exploring also novel integrative methods from the macromolecular (nm) to sub-cellular ( $\mu\text{m}$ ) levels. These include different kinds of phase space sampling, such as Brownian Dynamics (BD), advanced sampling methods and integrative methods that allow building structurally detailed 3D models of supra-molecular structures at cellular level. The workshop discussed all the major unresolved aspects that today represent the challenges in multiscale modeling. Established players in the field of multiscale modeling have led the discussion, and young scientists have productively engaged in the debate, with the aim of establishing new collaboration, advancing the field and developing new computational tools for the future of biophysics.

## 2 Major outcomes

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The workshop focused on major aspects in multiscale modeling: Time scale and system-size limits beyond the atomistic representations, integrative approaches including non-particle representations (surfaces, rigid shaped bodies, continuum models of solvent and membranes), and advanced/nonconventional configurational space exploration methods (i.e., beyond the current limits of Molecular Dynamics (MD): meta & accelerated MD), as well as the integration of these approaches. In the framework of mixed resolutions approaches U. Rothlisberger classified and highlighted the advantages and drawbacks of different QM/MM schemes, such as faster but less flexible "tightly-coupled" ones implemented in single dedicated codes, and the more general but less efficient "loosely coupled" implementation interfacing two codes. She

reported in particular about a newly developed interface that combines the advantages of both. H. Grubmüller reported atomistic studies of the ribosome, highlighting the use of atomistic simulations to refine cryo-EM data via flexible fitting. A. Rosa showed how the mechanistic action of the ribosome could also be tackled via coarse grained (CG) models. Simulation of diffusion and molecular association has also been central such as discussed by R. Wade who presented random Acceleration MD methods to compute relative residence time and introduced Brownian Dynamics (BD) to predict association rates. Recent advances in BD have been further discussed by G. Huber, who reported about the introduction of the flexibility in such simulations. J. Trylska reported on simulating the transport process of vitamin B12 and its conjugates with peptide nucleic acids. Her lecture discussed the development of ad-hoc force fields for peptide nucleic acids and highlighted the role of steered MD in the simulation of transport processes. A. Magistrato discussed the molecular assembly and mechanism of splicing in humans. W. Rocchia discussed emerging continuum-based models - based on electrostatics - to describe biological processes, such as chromatin formation, at the mesoscale level. CG model, possibly combining different resolutions, were discussed by P. Carloni who reported CG and mixed CG/MM simulations of G-protein coupled receptors (GPCR). Y. Miao presented the application of Gaussian accelerated MD simulations to capture ligand binding and the mechanism of association with intracellular factors, and V. Limongelli discussed the use of CG models and metadynamics to capture the dimerisation process. M. Cascella discussed a method to merge hybrid particle field molecular dynamics and CG models. S. Marrink reported about the new functionalities of the Martini force field for CG simulations, including polarizability. G. Voth presented a novel "ultra-CG" model with multi-internal states beads. The application of CG models has also been reported for the study of lipid droplets (S. Vanni, L. Monticelli), drug-drug interactions (T. Steinbrecher) and nanoparticles (G. Brancolini, M. De Vivo) Emerging and unconventional approaches have been reported by Z. Luthey-Shulten, J-P. Piquemal, A. Mulholland and C. Micheletti. Luthey-Shulten reported recent studies on the simulation of a minimal cell, including genetic and metabolic information. Piquemal reported how polarisable force fields are now reaching scalability on thousands of cores for simulating millions of atoms. Mulholland reported on the application of virtual reality to MD simulations, while Micheletti showcased theory and applications to the investigation of the formation of molecular knots. Predicting ligand on and off-rates in the framework of multiscale has also been discussed. B. Jagger reported about the SEEKER method that bridges BD and atomistic simulations to capture the kinetics of ligand binding.

## 3 Community needs

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The community needs have been discussed during three round tables of ~45 min each, which have been led by established scientists in the field. The first round table led by J. Trylska opened the discussion on several topics.

1. The accuracy of force fields (FFs) on the long time scales. This included the discussion on disordered proteins, whether analytical FFs are able to represent the configurational space and how experimentally assess it.
2. The community discussed the open issues in coarse grained (CG) simulations, in terms of what is missing in the representability (e.g. polarisation).
3. The community also identified the important need of introducing flexibility in Brownian Dynamics, to more realistically represent molecular association and diffusion processes.

The second round table led by P. Carloni focused on the challenge of simulating at atomistic level the cell and its components. The community converged on the need of adapting the current codes to the advances of the architectures hardware, envisioning a 5-to-10 years

challenge for the simulation of the cell. To reach higher scales, current state-of-the-art method must support coarse grained simulations. During the third round table, led by G. Voth, the community discussed about the importance of benchmarking free energy methods using blinded datasets, as done by the Drug Design Data Resource, and also touched on the importance of adding blinded methodological challenges for methods attempting to predict kinetics (noting here that data is more scarce), in order to truly create methods that are reliable. We also discussed the need for multiresolution approaches and the challenges of creating a theoretically rigorous 'one size fits all' coarse grained approach

## 4 Funding

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We have received support from Schroedinger and from the Swiss Industry fund (including Syngenta, Basf, Novartis, Roche). Future funding for CECAM workshop could also involve Psi-K and external funding sources, such as the National Institute of Health and National Science Foundation in the USA. While in Europe they can be provided by European Programs such as Horizon2020 e-Infrastructure or IMI projects in which computational modelling is an important component, and by the Swiss National Science Foundation in Switzerland. The Italian CECAM node could profit of Telethon, the Italian Cancer Research Association (AIRC), and the Italian Ministry for Education and Research (MIUR).

## 5 Will these developments bring societal benefits?

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For the first time in the history of computer modeling the size/time scales reached in silico by simulations start to come close to those obtainable in vitro, by experiments. Therefore contemporary computer scientists are in the unique position of directly checking the realism of their models, and on the other side, of giving accurate information about intra-cellular process where the experiment is impotent, predicting the behavior in new environmental conditions or of new therapeutic agents, therefore boosting in unprecedented way the biomedical research. This is even more important considering the fact that often there is no single experimental technique that can provide all answers for challenging large and complex biomolecular machines. The role of computations will be key to integrate available information to shed light in the mode action of those key components of the cell and generate new hypothesis to drive experiments. Increasing our knowledge of biomolecular function is of key importance in biomedicine, biotechnological, health and food applications. New societal challenges related to fighting emerging diseases will greatly benefit of an increased mechanistic knowledge of biomolecular function. This might in turn reduce costs associated with healthcare treatments. Novel drugs and therapeutic strategies may also help slow down the progress of the serious diseases, such as cancer, neurodegeneration, inflammation and emerging genetic diseases, which require profound knowledge of their mechanism of action. Computational methods and novel multiscale methods discussed in this workshop are perfectly positioned to impact the discovery of new drugs and therapeutic strategies, the development of better foods and new biomaterials, which ultimately will all contribute to an improvement of human health.

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# Avalanche dynamics and precursors of catastrophic events

Location: CECAM-FR-RA

Webpage: <https://www.cecarn.org/workshop-details/209>

Dates: Feb 04, 2019 - Feb 08, 2019

## 1 State of the art

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The idea of this workshop was to bring together international experts, in the field of avalanche dynamics, to discuss recent progress and insight from computational, theoretical and experimental approaches. This field is very challenging especially concerning modeling aspects, due to the computational difficulties related to critical slowing down of the dynamics and strong finite size effects. In the recent years, however, the computer power and new parallel coding techniques like GPGPU or large scale MPI programming techniques have allowed for significant progress in accessing reasonable time and length scales to address questions that were difficult to tackle before. Also on the experimental side, new approaches combining different techniques (e.g. various types of spectroscopy and rheology) allow for an investigation on a more mesoscopic scale of the complex dynamics, being able to probe scales that can be modeled within, for example, MD simulations. In addition, recent theoretical developments now address specifically the non-linear response of soft materials, providing new concepts and ideas to be tested in experiments and simulations. There seems to be a unique opportunity, at this point, in combining such efforts to develop a new understanding of avalanche dynamics, closely related to their technological applications.

## 2 Major outcomes

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The workshop was structured into nine thematic half-days plus three poster sessions. At the end of each day and some sessions we had detailed discussions to sort out the important open questions in the concerned fields. Abstracts and some of the talks (pdf and/or video) will be available on <https://avalanche2019.sciencesconf.org/>.

1. Non-universal features in avalanche dynamics.

This session focused on introducing the main reasons for non-universal features in avalanche dynamics, such as the presence of inertia and transient phenomena, where avalanches still play a dominant role for the overall dynamics. This first session also set the general theme of the conference within the different research areas on non-universal features and precursors of catastrophic events in avalanche dynamics.

2. Conditions for universality in avalanche dynamics.

Much effort has been invested to classify different systems exhibiting avalanche dynamics into universality classes regarding the underlying nature of disorder and the specifics in the interaction kernel (short or long ranged, strictly positive or with altering signs). This session aimed at clarifying the conditions under which one can expect universality and what should be the good classification criteria.

3. Precursors of catastrophic events.

One of the most urging questions to solve, also with respect to technical application, such as failure prediction and the forecast of extreme events in natural phenomena, is to find possible definitions of precursors of large events in the time series of

avalanches. This third session was be completely dedicated to this topic and introduced this general theme also for the discussions within the follow-up sessions.

4. Avalanches in transient dynamics.  
Some first works are now concentrating on avalanches in transient dynamics instead of being interested in the stationary state dynamics. This session concentrated on the specific role of initial conditions and parameter dependence on the avalanches in the transient dynamical regimes, a topic very important for applications where transient dynamics are ubiquitous.
5. Coarse grained models for avalanche dynamics  
The development of mesoscopic models, such as spring block models, mesoscopic elasto-plastic models, and various other coarse grained descriptions can help in understanding better the basic mechanisms for non-universal features in avalanche dynamics. This session focused on some recent advances in these modeling techniques and its application ranges.
6. Yielding transition.  
The yielding transition has been quite controversial with respect to its belonging in a specific universality class, especially the belonging to the depinning universality class has been shown to be questionable. Depending on the observables considered and the dynamical regime of interest the reasoning can be very different. This session helped to sort out the necessary numerical (and experimental) tests needed to converge the understanding of this strongly debated subject.
7. Depinning transition.  
The depinning transition is probably the best understood dynamical transitions exhibiting avalanche dynamics. But this workshop helped to turn the focus on the study of transient dynamics and situations with non-universal dynamics that had not been strongly addressed in this context.
8. Instability in solid earth systems.  
Geophysical systems with avalanche dynamics, such as snow avalanches and earthquakes are related to very similar questions compared to the depinning and the yielding transition and this session helped to isolate common questions that can be tackled in a common framework.
9. Other systems exhibiting avalanche dynamics.  
Beyond the above examples for systems exhibiting avalanche dynamics there is a myriad of systems dealing with strongly correlated intermittent dynamics, as is the case for example in Martensites, in foams and even in active matter systems. This session made connections to these different fields.

## 3 Community needs

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The CECAM workshop on avalanches and precursors to catastrophic events was a success and has clearly exposed the need of meetings, where researchers working on the diverse aspects of out-of-equilibrium critical dynamics and intermittency can exchange their ideas and put their work within a broader context. All participants have expressed their satisfaction regarding the present workshop. It was highlighted that the choice of the focus was timely and excellent to create and strengthen synergies between experiments and computational approaches. It was suggested to be highly desirable to continue a series of European workshops on avalanche dynamics to promote the development of unifying concepts and stimulate further collaborations between researchers exploring the behavior of metastable systems in simulations and those exploring them in experiments. All participants were satisfied with the set of topics covered by the workshop, that appeared sufficiently diverse without losing a coherent focus in the discussions. For future meetings, it was suggested to include related

themes such as intermittency in biological systems, like for example neural networks. Some of the participants felt that discussions in possible future meetings would benefit from more contribution from analytical theories, and we believe that further efforts to integrate theoretical approaches, simulations and experiments on avalanche dynamics and the possible precursors to catastrophic failure would fit perfectly CECAM workshops scope and mission.

## 4 Funding

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This event was cosponsored by CECAM-RA (6 k€) and CECAM-FI (9 k€). We applied to two CECAM nodes to ask for a larger budget, since the duration was twice as long (5 days instead of 2,5) and the number of participants twice as large (70 instead of 35) as compared to usual meetings. Knowing about the application to two nodes, CECAM-RA unfortunately reduced significantly the usual budget attributed, which put us in a difficult situation at first. In the end the CECAM-FI node provided help and we were sponsored additionally by several local funding agencies: ENS de Lyon - 6 k€ Region Auvergne-Rhone-Alpes - 4,8 k€ Fondation nanosciences (Grenoble) - 4 k€ UGA-INP (Grenoble) - 2 k€ LabeX OSUG (Grenoble) - 4,8 k€ We used the budget to waive the inscription fees of all participants and to pay travel for invited speakers and participants in need of support. Another possible funding for future meetings of this kind (foreseen in two years from now) could be the Maupertuis program.

## 5 Will these developments bring societal benefits?

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Critical dynamics, intermittency and the search of precursors to catastrophic failure is crucial to a wide range of technologies, e.g. in the dynamics of domain boundaries in micro- or nano-scale devices using ferro-magnetic or ferro-electric materials. On a larger scale, technologies involving amorphous and crystalline materials, ranging from bulk metallic glasses, crystalline micro-pillars, to large-scale construction materials and granular matter involve many questions about plasticity and sudden yielding. Further applications can be found in the design of flow properties of soft glassy materials such as gels, pastes and foams. And when thinking about even larger scales, these type of instabilities play a major role in the determination of risks and the installation of warning systems in earth science problems, such as for earthquakes, snow avalanches and landslides. The workshop brought together a wide spectrum of scientists from different domains and the discussions demonstrated the need to develop novel fundamental understanding and complementing experiments with computational approaches to advance material design, process optimization as well as warning systems. The research discussed in the Workshop has the potential to help develop new products or optimize and enhance existing ones; optimize industrial processes to achieve higher sustainability; enhance quality of life through better designed products, industrial processes and better warning systems for natural catastrophic events. In addition, creating such an environment in which academic scientists communicate across disciplines helps enhancing integration of scientific culture and communication with societal needs, with new opportunities to develop mixed academic-industrial collaborations and to access diversified funding.



## 6 Participant list N/A

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### Organizers

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# Correlated electron physics beyond the Hubbard model

Location: CECAM-DE-MM1P

Webpage: <https://www.cecam.org/workshop-details/182>

Dates: Feb 04, 2019 - Feb 08, 2019

## 1 State of the art

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Realistic modeling of materials with strong electron correlations presents one of the central challenges in condensed matter theory to date and hold promises for novel materials developments in the domains from materials for energy applications to novel high temperature superconductors and topological states of matter. On the theory side, the Hubbard model is one of the central models to understand various aspects of strongly correlated electrons. However, links between the Hubbard model and real materials can be ambiguous and the Hubbard model can even qualitatively fail to describing real materials whenever non-local interactions are not efficiently screened. Various theoretical approaches from quantum physics and chemistry communities including Quantum Monte Carlo approaches, coupled cluster theories, Configuration interaction expansions, embedding theories, extensions of dynamical mean field theory, as well as renormalization group approaches and diagrammatic perturbation theory have been put forward to address strong electron correlations in presence of non-local interactions. However, our understanding of the electronic structure of these systems is still rather limited as regards electronic phase diagrams, excitation spectra, and excited state dynamics - both on the pure model side and particularly when it comes to modelling real materials. The development of next-generation realistic many-body computational tools which are fast, reliable, and able to describe non-trivial quantum states of real materials requires clarification how non-local interactions affect the electronic properties of correlated electron systems. The workshop aimed to foster a clear understanding of merits and shortcomings of different simulation methods currently under development by bringing together the respective scientific communities from physics and chemistry and set the stage for developing novel simulation tools for real materials featuring correlated electrons.

## 2 Major outcomes

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The program consisted of 26 invited talks of 40 minutes (30+10) each, one poster session with 29 posters and 3 round table discussions of 45 minutes each. In addition, social events (reception and conference dinner) allowed for informal exchange. The invited talks were given by well-established scientists from different theoretical communities, which acted as platform for interesting cross-/interdisciplinary discussions. The invited talks were followed by a poster session where the young researcher participants showed their scientific work and exchanged ideas with each other and the present experts in the field. The organization was very compact with the participants accommodated in the same hotel fostering exchange and discussion also outside the meeting room. The central issue standing out in this conference was how to advance the field in order to achieve reliable predictions of properties of materials hosting strong electronic correlations. The conference gathered experts in a diverse collection of methods suitable to tackle this task, which was the foundation for lively discussions about both fundamental issues of the field and technical details of novel methods. On the one side, experts

in methods designed for low energy methods presented and explained details of their respective advances, e.g., in dual approaches (E. van Loon, A. Lichtenstein), different Monte Carlo approaches (J. LeBlanc, L. Wagner, S. Wessel, B. Svistunov, N. Prokofiev, S. Sorella), renormalization theories (S. Andergassen, C.-W. Tsai), and variational approaches (R. Arita, G. Booth, M. Schüler). On the other hand, methods designed to tackle realistic materials including long-ranged interactions from the beginning were presented, like GW based methods and extensions thereof (P. Werner, J. Lischner, S. Biermann, Ferdi Aryasetiawan, Mark van Schilfgaarde), and density functional based methods (M. Cococcioni). Finally, we had experts in the field of downfolding methods (i.e., the task to excerpt low energy models from a high-energy manifold based on ab-initio methods) talking about the subtleties and advances in this field (M. Rösner, Carsten Honerkamp). Multiple speakers took the opportunity to present applications of this zoo of methods to real materials (A.-M. Tremblay, A. Millis, S. Biermann, F. Aryasetiawan). The round table discussions revealed following issues: First, to assess the multitude of available methods applicable to model systems, a common benchmark is quite desirable. Often these methods are complementary and best applied to different regimes. Here, a benchmark on common grounds against numerical exact results from, e.g., diagrammatic Monte Carlo simulations for a non particle-hole symmetric, slightly doped one-band model with local interactions on the order of the bandwidth and non-local interactions is highly desired and would highlight if certain approximations, for instance the locality of the self-energy or the neglect of vertex corrections to non-local properties, hold. In a second step, it is unclear how this relates to real materials: due to the complexity of real materials, a numerical benchmark will not be possible in the next few years. Here it was brought up, to which extent experiments are a useful benchmark for methods only applied to model systems. One suggestion are artificially engineered ad-atom systems closely resembling model systems. Besides experimental verification, the importance of comparing results from complimentary methods was brought up. Another important topic in the discussions were different downfolding techniques and their reliability. Here, the advances of the constrained functional renormalization group theory approach shed light on the applicability of computationally more lightweight and therefor more often used constrained random phase approximation. However, it is unclear how these findings apply to real materials. It is now planned to perform a benchmark for the case of graphene.

## 3 Community needs

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As discussed above, the advancement of theories of the operation of strongly correlated electron systems requires the development of novel theories and codes which can i) deal with the complexity given by electronic correlations in model systems, ii) treat realistic materials which include many bands, not all of which may host correlation effects, and iii) combine ab-initio and model approaches in an optimal way. The development of such theories and the resulting computer software will benefit the broad community of theoretical researchers, but also have important impacts on experimental studies. Here, the interplay with experimental researchers is crucial to provide excellent data sets which can serve as benchmarking and ultimately lead to a fruitful interplay between theoretical predictions and experiment. However, to achieve this, a continued investment is required, as method and code development usually occur on a longer time scale compared to the study of applications. This also requires the training of masters and PhD students not only in physics, but also in computer programming (including parallelization of software) and use of high-performance computing resources. We have to work cross disciplinary to tackle correlated electrons (benchmarking; gaining predictive power) which requires networking and collaborations. Here, networks like Psi\_k and coordinated research programs bringing modelling experts together have proven extremely useful.

## 4 Funding

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We could use the opportunity of this meeting to discuss among the attendants from German speaking countries, about a new collaborative initiative (DACH project by Deutsche Forschungsgemeinschaft (DFG), Fonds zur Förderung der wissenschaftlichen Forschung (FWF) and Schweizerischer Nationalfonds (SNF)) for modelling systems with strong non-local correlation phenomena including topological phenomena. Additionally, we could strengthen and foster collaboration between researcher groups from European based projects and the US based Simons Collaboration.

## 5 Will these developments bring societal benefits?

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Our workshop contributes to the potential development of technologies based the unique properties of materials hosting strong electronic correlations. This may have a number of societal and economic benefits, especially in terms of: - The advent of superconducting materials with record high critical temperatures, leading to cheaper energy transfer and lower cost for cooling electromagnets based on superconducting materials. - Novel switching devices utilizing the large susceptibilities in correlated materials, e.g., Mott metal-insulator transition. - Using properties of materials with colossal magnetoresistance as memory devices. In general, the study of strongly correlated materials is very much basic research, such that direct applications cannot be foreseen to full extent at this point. However, it is at the heart of science, that basic research pays off in one way or the other. Especially the development of new methods and training of students has a strong indirect economic and societal impact.

# 6 Participant list

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# From sequences to functions: challenges in the computation of realistic genotype-phenotype maps

Location: CECAM-ES, University of Zaragoza

Webpage: <https://www.cecarn.org/workshop-details/197>

Dates: Mar 13, 2019 - Mar 15, 2019

## 1 State of the art

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How genetic variation contributes to phenotypic variation counts amongst the most relevant questions to understand the evolutionary process. The experimental characterization of the genotype-phenotype (GP) relationship is a formidable and expensive task; therefore, and together with models of variable complexity and realism, computational approaches have been used since long to make predictions of phenotypes from genotypes and to uncover the statistical features of that relationship. Important advances notwithstanding, we face an apparently unsurmountable problem: the astronomically large size of the space of genotypes. The motivation of this workshop was to explore the state-of-the-art, to discuss limitations of current approaches and to synthesize the perspectives of the topic. An important starting point put forward by P. Hogeweg was whether dynamics on GP maps can be studied straight forward, or whether, instead, the GP map is a highly dimensional and complex object that has a dynamics in itself. This is an unsolved question of relevance, among others, to characterize evolutionary innovations. Also, L. Altenberg raised the question of understandability: perhaps the relation between genotype and phenotype cannot be disentangled; maybe there is no possible hierarchical decomposition of the problem, such that we can see it working (and perhaps “make it work”) but cannot understand it. The probability of expressing different phenotypes or of experiencing mutations that modify the current phenotype depends, beyond a possible selection for evolvability, on the intrinsic structure of the GP map. In his contribution, A. Louis discussed the roots of what is called phenotypic bias, and argued that simpler outputs are much more likely to appear than complex ones from an information theoretic viewpoint. Since the GP map cannot be systematically studied by experimental means, a number of well-motivated models have been proposed and studied through the years. Prominent examples are the RNA sequence-to-secondary structure map and the hydrophobic-polar (HP) model for protein folding. Two of the participants discussed the latest advances on computational and algorithmic research of those two models: J. A. García-Martín summarized his research on the exhaustive enumeration of GP maps and introduced the idea of the dual partition function, a technique that could be instrumental to perform unbiased samplings of structure spaces in RNA; A. Irbäck summarized current knowledge on the HP model and the limits of exhaustive enumeration. Other, more recent, proposals of models for GP maps include metabolism based on a set of reactions, gene-regulatory networks, the quaternary structure of proteins or the construction of metabolism from genomes through different expression levels. In her talk, N. Martin presented a new map, the so-called Dawkins’ biomorphs, showing evidence of general features shared by other maps. Together with advances in the quantitative characterization of natural relationships between sequence and function, computational and theoretical results support that GP maps may have universal architectural properties. Some of those properties can be used to derive relatively simple prediction of phenotype size, for example, as M. Weiss discussed in his presentation. However, and despite a number of formal efforts, the way in which the structure of GP maps affects adaptation is a poorly explored question due to the intrinsic difficulty of obtaining sound quantitative results in biologically realistic genotype

spaces. In his talk, however, P. Catalán discussed the effects of phenotypic bias in a recent, multi-level GP map called toyLIFE, and demonstrated that emerging phenotypes select for most abundant solutions in genotype space. In his turn, J. Krug explained how the accessibility of alternative phenotypes and the structure of populations changes when, instead of simple point mutations, recombination is considered. Related to evolutionary dynamics, J. Aguirre discussed how the network-of-networks structure of genotype spaces entails sudden adaptive transitions. Finally, B. Khatri discussed thermodynamics of evolutionary dynamics on genotype spaces, a promising approach that takes advantage of well-known techniques in physics. Establishing the strengths and limitations of simplified GP maps should improve their application to the prediction of function for natural sequences (as RNA or proteins), and it might be relevant as well in biotechnology and systems chemistry approaches. We enjoyed a number of talks where the relationship between formal concepts and empirical findings was deeply discussed. S. F. Elena discussed the concept of fitness landscape and its properties in the light of our knowledge of RNA virus populations, highlighting the important gap still existing between empirical data available and whole genotype spaces. M. Tarnowski presented the strengths and weaknesses of current technologies to empirically sample genotype spaces. J. L. Payne discussed how the functional spaces of short RNA sequences can be experimentally characterized, and A. Vilorio-Cano summarized through an empirical example how mutation bias affects evolutionary dynamics. Finally, the way in which generic properties of the GP map shape adaptation in an ecological context have rarely been formally explored so that, as of today, the overarching question of whether organismal phenotypes can be derived from microscopic properties of genotype spaces remains open. In this respect, R. Díaz-Uriarte discussed limitations of models of cancer progression based on driver, sequential mutations, which take into account neither the complex topology of realistic fitness landscapes nor the many levels that go from genotype to phenotype.

## 2 Major outcomes

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All contributions by participants (briefly summarized in the former section) highlighted their more recent research on the topic and pointed out to multiple open questions of their interest. During the discussion sessions, we managed to extract three major avenues for future research which include as subtopics different aspects. These three avenues have a transversal character in that advances in knowledge will be foreseeably gained through an interaction between theory, computational research and new empirical results. The three main topics are as follows.

1. Evolutionary relevance of phenotypic bias the skewness in the relative abundance of different phenotypes has been recently quantified. It turns out that entropic effects may condition the emergence of certain phenotypes. A future advance, which is now close to be quantitatively settled, is the relative importance of entropy versus fitness in evolution. It was pointed out that a formal framing of this question would probably solve the neutralist versus adaptationist discussion. This is one of the main contributions that the study of GP maps might add to current evolutionary theories. The existence of phenotypic bias is also of relevance for synthetic biology and design through directed evolution. Up to now, in vitro design of molecules with specific activities has ignored entropic effects. However, given the intrinsic evolutionary instability of small phenotypes, it is likely that selection for robustness (in the form of large phenotypes) should be added as a design requirement. There are two issues that have to be kept in mind and still require further research before the evolutionary relevance of phenotypic bias can be fully understood. They are the levels of epistatic interactions, which deeply determine the structure of fitness landscapes and the accessibility of



phenotypic solutions, and the structure of genotype spaces when multiple mutational mechanisms are considered.

2. Organisms as a result of connecting levels from genotype to phenotype. Most of the GP models we currently use consider a single level (from sequence to structure, for example). However, organisms result from the expression of genotypes through several levels of organization and expression. Currently, we do not have models for whole organisms. In this respect, we discussed how one could generalize beyond the current level of description, and which would be the difficulties when the different levels cannot be formalized as sequences. This is relevant in the interpretation of data, since intermediate levels have to be inferred. Beyond theoretical and computational issues, this topic is important from a very basic viewpoint: Is decomposability (in a number of levels) possible at all? Feedbacks between levels might be essential in evolution. Concepts such as universality, which to a good extent seems to hold in different GP maps, might come to help. Commonalities among GP maps can likely be exploited to reach valid, general results. In this respect, the limits of computation in the analysis of GP maps must be aided by the further development of theoretical approaches.
3. Many-to-many GP maps and environment. Despite the fact that most models stick to the many-to-one representation of GP maps, the latter are actually many-to-many: many genotypes code for the same phenotype, but also every genotype might express multiple phenotypes, especially depending on the environment. Even in the same environment, phenotypic promiscuity (different phenotypes beyond the most common one are expressed with non-zero probability) is an important adaptive mechanism. Our conclusion was that at least three aspects have to be included in future models: promiscuity/phenotypic plasticity, seascapes (representing variable environments) and overlapping genotype networks. There might be important evolutionary consequences, such as the possibility that valleys (which in a simple framework for evolution trap populations) are not such when the environment changes. Also, one should consider the evolution of GP maps versus the evolution on GP maps, since evolutionary innovation might be controlled by the former.

## 3 Community needs

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If studying the relation from genotype to phenotype aims in any way to be useful, it must face the problem of dealing with hyper-astronomically large samples of sequences. Even sequences with lengths below that of typical functional proteins imply sequence spaces of size larger than the number of particles in the universe. Therefore, though computational approaches have importantly guided our understanding of GP maps, many questions cannot be solved using current techniques and conceptual frameworks. We have discussed two possible alternatives. One is the future use of quantum computing. This avenue is highly speculative at the moment, but will be kept in mind. The second one is to attempt a sort of inverse design of the problem, that is, to formulate specific questions based on theoretical predictions or empirical inferences that can be computationally tested with the available infrastructure. Still, the question of how far we can get by means of numerical explorations of the problem remains an open question. One of the concerns we discussed was the way in which other scientific communities could be reached. Indeed, we believe that many of the results we have gathered and their implications are of relevance for communities (especially in evolutionary experimental biology) that approach their topics in a very classical fashion, meaning that they mostly use concepts that were put forward in the first half of the 20th century—in the framework of the New Evolutionary Synthesis. These concepts were developed before the sequencing era, and therefore do not incorporate, in general, any of the new advances contributed by this technique. To this end (among others) we have decided to

write a joint “Perspectives” paper that summarizes the most recent advances in the field and puts emphasis on the applications and implications of current GP map understanding. We have contacted Dr. Tim Holt, editor of the Journal of the Royal Society Interface, and he replied that they would be happy to consider such a contribution. We have set an expected submission date by September 2019, and CECAM will be informed of our progress—and properly acknowledged in the paper for its generous contribution to make this workshop possible. Regarding event organization, we have pointed at the possibility that a similar meeting could be organized in two-three year time, and we would gather specific funding—perhaps again in the framework of CECAM under the leadership of a different country.

## 4 Funding

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Up to now, typical funding channels of the topic have been national funding schemes and, occasionally, European consortia. The latter, in general, have not focused specifically on the study of GP maps, but have the topic of our meeting as part of a larger question of interest (such as the evolution of evolvability or the emergence of evolutionary innovations). It is important to remark that research on GP maps has been until recently a relatively cheap endeavor, since most approaches were either theoretical or computational (using already available resources). We discussed the possibility of applying for a project within the new Quantum Flagship initiative of the EU in the near future, though first we need to clarify the details of the quantum implementation of the problem. A large-scale study is not yet viable given the state-of-the-art of quantum computing, but GP maps could be in all likelihood one of the open scientific problems that could immensely benefit from further developments of the field.

## 5 Will these developments bring societal benefits?

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The community that met in this workshop mainly works on basic science, even if their backgrounds covered from computer science to biochemistry, and even if some of them carry out original experiments in the area. Therefore, most of the benefits of future developments will affect the basis of our knowledge of evolution. This contribution by itself is however very relevant. There is certain implicit consensus regarding the limitations of current evolutionary theory, which stem from a number of assumptions that do not have empirical support. A new theory must emerge from the integration of results on deep sequencing of molecular populations and a profound understanding of the structure of GP maps, and this theory should substitute in the mid-to-long run our current formulation of evolutionary dynamics. Still, the potential benefits of this foreseen change of paradigm do not stop here. As mentioned, we believe that synthetic biology should incorporate new concepts to yield new useful solutions in any of the areas it is involved (drug design, cellular control, development of microorganisms with environmentally relevant functions, etc.). Also, all applications of evolutionary thinking to societal problems (e.g. viral evolution and disease control, emergence of new diseases, response of microbiomes to changing environments) can potentially benefit from a deeper knowledge of GP maps. The fact is that most information is obtained at the level of genotypes, but changes in genotypes affect their expression—that is, phenotypes. There is a tremendous lack of knowledge of how mutations are eventually expressed in organisms, and ours is—we believe—one relevant way of advancing in this respect. So, even if this community is not

working on the immediate applications of the field, it is aware of its relevance in applied topics, and concerned about foreseeing possible applications.

## 6 Participant list

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### Organizers

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# Improving the theory in density functional theory

Location: CECAM-HQ-EPFL, Lausanne, Switzerland

Webpage: <https://www.cecam.org/workshop-details/196>

Dates: Mar 18, 2019 - Mar 22, 2019

## 1 State of the art

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The balance between low computational cost and useful (but not yet chemical) accuracy has made density functional theory (DFT) and its time-dependent formulation (TDDFT) a standard technique in most branches of chemistry and materials science. DFT is useful, in fact, for a dazzling variety of electronic structure problems. Furthermore, it is increasingly applied beyond its traditional scope and validity in order to solve electronic structure problems under extreme conditions. However, DFT has many limitations: it requires too many approximations, it presents failures for fundamental gaps and strongly correlated systems, it is too slow for complex systems. In the following, we give a concise compilation of some these limitations.

Exchange-correlation (XC) functionals in DFT Early approximations such as the local density approximation (LDA), generalized gradient approximations, and hybrid functionals have led to a success story. However, further progress in deriving more accurate approximations for a wider scope (such as incorporating long-range van der Waals interactions or curing the self-interaction error) has proved increasingly difficult. This has led to more empiricism in functional construction and, eventually, to a plethora of available approximations. Notwithstanding recent progress in developing more sophisticated approximations such as meta-GGAs, non-local functionals for van-der-Waals interactions, range-separated hybrids, or functionals based on the RPA, a clear route for advancing the field does not exist. A striking example of the limitations of the current theory is the metallic KS ground state of strongly correlated Mott insulators. There has been some (limited) progress in the density functional description of strongly correlated lattice models, but it is unclear how to turn this insight into a practical density functional scheme for strongly correlated systems. Beyond its traditional scope, DFT has recently been applied to warm dense matter (WDM). DFT simulations of WDM melt properties and thermal conductivities are crucial for achieving nuclear fusion under laboratory conditions. Furthermore, DFT simulations of WDM can elucidate fundamental questions in planetary sciences, such as the formation processes of planets and the search for exoplanets. The main caveat in these simulations is the lack of XC approximations that properly account for temperature effects.

Exchange-correlation functionals in TDDFT TDDFT has been successfully applied in predicting excitation spectra of molecules and solids in the linear response regime. Also, more complex time-dependent phenomena, such as matter in strong laser fields and optimal control of electron (spin) dynamics are continuously being explored through TDDFT. As in static DFT, the greatest challenge is the derivation of accurate approximations to the XC kernel. Currently the standard approach is the adiabatic LDA, where the ground-state LDA functional is evaluated on time-dependent densities. Present limitations and further developments are needed in charge transfer excitations, double excitations, excitons, and alternative approximations to the XC kernel from many-body theory.

## 2 Major outcomes

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The aim of this workshop was to advance the field of DFT and TDDFT. It focused on (1) new developments for the construction of exchange-correlation functionals in DFT and TDDFT and (2) alternative methods such as many-body perturbation theory and reduced density matrix functional theories which can enrich the theoretical framework of DFT and, thereby, improve upon its accuracy and range of applications. A brief survey on the major outcomes of this workshop is given below.

**New developments on exchange-correlation functionals in DFT:**

- Several new developments regarding fundamental progress extending the general scope of DFT were presented. In his presentation "On the Constrained-Search Formulation", Mel Levy summarized the constrained-search formulation and gave an overview of its implications for modern electronic structure theory, in "Density functional theory without density functionals" Andreas Savin discussed the possibility of circumventing the solution of the Kohn-Sham equations, in "Accurate treatment of exchange vs self-interaction corrections in the KS equations" Nikitas Gidopoulos presented progress on correction of the self-interaction error, in "Fractional Perspectives of DFT, with Quasiparticle and Excitation Energies from Ground State Calculations" Weitao Yang formulated a possible route to excitation energies within DFT as an alternative to TDDFT, in "Density-functional theory of quantum phases and their dynamics far from equilibrium" Hardy Gross formulated a general framework for the DFT of quantum phases such as magnetism and superconductivity, and Kieron Burke presented a practical framework to analyze errors in DFT calculations by separating density-driven and functional-driven errors.
- Several recent developments on exchange-correlation functionals based on iso-orbital indicators were presented such as "Density-gradient-free variable in exchange-correlation functionals for detecting inhomogeneities in the electron density" by Fabien Tran, "An exchange-correlation functional for bulk, surface, and confinement physics" by Attila Cangi, "Accurate charge-transfer and improved band structures from a new meta-GGA" by Stephan Kümmel.
- Recent progress on range-separation of exchange-correlation functionals was presented such as "Electronic and optical excitations from screened range-separated hybrid density functional theory" by Leeor Kronik,
- Recent progress on the construction of exchange-correlation functionals with an explicit dependence on temperature were presented by Aurora Pribram-Jones in "New Approaches to Temperature-Dependent Density Functional Approximations"

**New developments on exchange-correlation functionals in TDDFT:**

- Developments beyond the adiabatic approximation were discussed and presented, such as "Time-dependent density-functional theory: thoughts about the future" by Robert van Leeuwen, "Nonadiabatic Dynamics in Single-Electron Tunneling Devices with Time-Dependent Density-Functional Theory" by Nicole Helbig, "Beyond the Adiabatic Approximation in TDDFT" by Neepa Maitra, and "Density-matrix coupled time-dependent exchange-correlation functional approximations" by Lionel Lacombe.

**Recent theoretical progress extending the scope of DFT and TDDFT:**

- The utility of many-body perturbation theory, reduced density matrix functional theories, and open quantum systems were major outcomes of the presentations "Hierarchies of methods towards the exact Kohn-Sham correlation energy based on the adiabatic-connection fluctuation-dissipation theorem" by Andreas Görling, "Non-locality in space and time in functional theories" by Lucia Reining, "Connector approach for electronic excitations" by Marco Vanzini, "Wigner density functional theory" by Carlos L. Benavides-Riveros, and "Unravelling open-system quantum dynamics of non-interacting Fermions" by Roi Baer.

## 3 Community needs

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While density functional theory (DFT) is being applied successfully in ever more areas of science, the theory behind it is rarely covered at sufficient depth at international conferences which are usually dominated by a plethora of applications in specific fields. The unique aim of this conference is to provide an interactive venue for DFT developers for discussing (1) the theoretical developments rather than computational methods or specific applications and (2) the current theoretical challenges DFT is facing. The ultimate goal of this conference is to identify methods and approaches needed to increase the accuracy and efficiency of the current state-of-the-art and hence make DFT systematic and reliably applicable beyond its current scope. The need for this kind of workshop which focuses on theory and methods development is further stressed by the feedback we received after the workshop from both participants and invited speakers. Below we give some excerpts of the feedback we gathered:

- Excellent selection of topics and balance between more senior and more junior speakers.
- Ample time for discussion, both formally and informally.
- Extremely effective format, with 35 minute talks + 25 minute discussions associated with each invited talk, and 20+10 for contributed, meant that a much larger fraction of participants were actively involved for much longer each day than in usual workshops.
- Junior and senior scientists were inspired to contribute to the discussions.
- Discussions were extremely interesting, engaging and constructive.
- Very friendly atmosphere throughout the workshop.
- Perfect schedules for the whole day, and the breaks, and refreshing snacks were greatly appreciated.
- All in all, one of the best workshops I have ever been to, from all aspects, scientific and social.
- Good selection of participants: Different fields were covered and enough diversity present to be inspiring, yet close enough so that one could really talk to one another.
- Very active discussions, encouraged by the format. 35 minutes is a good length for an invited talk and it was good to have "special" and "general" discussion time.
- Right size of the workshop. Quite a few people there, but a limited number of people so that one could really talk and discuss.
- Great that so many of the well-known experts were there from beginning to end.
- Format and schedule of the workshop were perfect, in particular the long time for discussions.
- The selections of speakers/participants was very well chosen resulting in a program well focused on the title of the workshop.
- The workshop was characterized by very lively and enlightening discussions throughout the whole time.
- In summary this was one of the best CECAM workshops I have ever been, meetings like this workshop that are well focused and enable intense discussions and interactions between the leading researchers in a field are most productive.
- Size was ideal for a workshop of this nature: A relatively small, but full room.
- Most people at the workshop cared about similar issues but brought different perspectives, so discussions were lively and interesting.
- The format was excellent. Most talks had a full hour slot. Having to finish at 35 minutes left plenty of time for in-depth discussions.
- Perfect balance of lecture-time / question-time / coffee breaks.
- Also optimal length of time: 5 full days.
- Ideal location.

There is something about this CECAM location that creates a very enjoyable atmosphere. I don't know if it is because of the beautiful surroundings, or the enjoyable walk from the hotel, the coffee-room upstairs, ... but there is something at this location that makes people get into a good mood, which translates into fun, interesting conversations.

## 4 Funding

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The funding for this workshop came from CECAM and the Max Planck Institute of Microstructure Physics, Halle, Germany. The discussions during this workshop were primarily focused on the science. The identification of funding channels supporting the research of this workshop were not a topic of general discussion. However, this is a point which could be stressed more and possibly be included in a future workshop of similar kind.

## 5 Will these developments bring societal benefits?

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Density functional theory has become one of the most widely used computational method in chemistry, condensed matter physics, and materials science. It enables first-principles predictions in an incredible scope of applications such as the search for efficient and inexpensive catalysts, materials for batteries with higher energy-storage capabilities, more efficient materials for solar cells, materials for superconductivity at room temperature, understanding of the equation of state and transport properties of matter under extreme conditions. This is just a small sample of the scope where DFT is incredibly useful in terms of technological applications. Theoretical developments and progress in DFT, therefore, have an enormous potential to both impact and transform key aspects of industry, economy and society.

## 6 Participant list

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### Organizers

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**Burke, Kieron**

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**Tran, Fabien** - Vienna University of Technology, Austria  
**Ullrich, Carsten** - University of Missouri, United States  
**Van Leeuwen, Robert** - University of Jyväskylä, Finland  
**Vanzini, Marco** - EPFL, Switzerland  
**Wasserman, Adam** - Purdue University, IN, United States  
**Yang, Weitao** - Duke University, United States



# Big data and uncertainty quantification: statistical inference and information-theoretic techniques applied to computational chemistry

Location: 26-29 March CIB, room GA 3 21, 1-3 April CECAM, room BCH3113

Webpage: <https://www.cecama.org/workshop-details/173>

Dates: Mar 26, 2019 - Apr 03, 2019

## 1 State of the art

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Generally, the CIB semester programme *M3+C3: Multi-scale Mathematical Modelling and Coarse-grain Computational Chemistry* is built around a central core of applied mathematics as it concerns molecular and materials modelling from atomistic to continuum scales. More specifically, the workshop that took place on the CIB and CECAM premises, was concerned with the interplay between analytical (“bottom-up”) and data-driven coarse-graining methods, and discussed ways to systematically use effective dynamics for performing system tasks such as estimation, filtering and prediction, in a data-driven framework, with a special focus on statistical and Bayesian inference techniques. An important recent development that was discussed during the workshop, was the systematic use of data in the derivation and validation of the effective dynamics, especially in order to correct misspecification or uncertainty in the fine-grain (“root”) models. In data-driven approaches this is often done by means of suitable loss or discrepancy functions that measure the difference between the output of a candidate coarse-grain model and the available data. That said, the core topics of the workshop ranged from Bayesian parameter estimation and model calibration to sensitivity analysis, kernel-based statistical methods and novel approaches such as machine learning or sparse learning. The format of the workshop featured a rather dilute programme, with very few talks, lots of time for informal blackboard discussions and (Oberwolfach style) discussion sessions, during which certain topics that came up during the presentations were discussed in a broader context. (For example, it turned out that machine learning and sensitivity analysis were such topics.)

## 2 Major outcomes

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Key aspects that were discussed during the workshop were (making no claims of being complete):

1. Parameter estimation in a semi-parametric framework: When given noisy data from some microscopic dynamics (projected onto a set of pre-selected coarse variables), estimating parameters in the effective molecular dynamics, or even the functional form of the equations is known to often suffer from a systematic bias under too-fine sampling. During the workshop, various remedies have been discussed to solve the problem, among which were sparse sampling (Zhang, Pantazis), semiparametric approaches (Krusmscheid), Bayesian inference (von Toussaint, Zygalakis, Di Blasio), or kernel-based methods (Duncan).

2. Global sensitivity analysis: Variance-based sensitivity indices, such as Sobol indices are a popular tool to assess global sensitivities in a dynamical system. However as first-order indices, they may provide misleading information about the the relative and absolute importance of certain parameters, especially in cases when there are many, as has been pointed out by various authors (e.g. da Viegua, Knio). Accordingly, several authors have devoted their talks to refined sensitivity analysis based on concentration inequalities (Wang), polynomial chaos expansions (Le Maître), or statistical learning ideas (Wu).
3. Prediction & criticality: Depending on what aspect of a molecular system is of interest, one often has at one's disposal various different coarse-grain models with very different predictive power. It has been critically discussed that most of these models reproduce equilibrium properties rather well, but may fail to predict bifurcations or noise induced conformational transitions. This aspect has been critically discussed during the workshop, with regard to capturing metastability (Pavliotis, Pinski, Lelièvre), nonreversible and Markovian approximations (Sharma, Neureither), or the usefulness of machine learning techniques (Zabaras, Fass, Wu).

## 3 Community needs

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To summarise some of the discussions during the both weeks of the workshop, a common theme was that many techniques used in coarse-graining, parameter estimation or sensitivity analysis rely on assumptions of stationarity, thermodynamic equilibrium or the like that may cease to exist in more realistic settings. On the other hand, some of the novel approaches based on statistical learning or machine learning do not rely on such stringent assumptions, but they are typically ignorant of the underlying physics, even though there are many cases in which they produce remarkable results. It appeared during the workshop that the interplay between these different aspects (e.g. statistical vs. physics-informed coarse-graining) still needs to be critically discussed and that the underlying algorithms need to be better understood---especially when these algorithms work, why they work and why they sometimes do not work. Interdisciplinary CECAM workshops like this one are a good opportunity to discuss these pressing issues and to agree on suitable benchmark systems.

## 4 Funding

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We had some contributions that were coming from speakers with a more applied or industrial background. Even though we believe that most of the scientific problems discussed during the workshop were quite fundamental, the contributions from this side were quite substantial and valuable. Therefore, we believe that the H2020 COFUND postdoc programme may be a suitable funding scheme to address some of the problems discussed during the workshop, if the industrial participation is not too dominant. Long-term programmes such as the one at the Bernoulli Center, the Institute Henri Poincaré or the Simons Foundation may be a good format too.

## 5 Will these developments bring societal benefits?

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The interplay between statistics, computational engineering and statistical physics had been very fruitful, in that many applications were discussed from various different standpoints (e.g. model uncertainty, computational complexity, predictive power). Application-driven problems that were discussed by many participants and that we believe to be relevant from an industrial and societal perspective were e.g. chemical reaction networks, with applications to combustion or plasma physics, or weakly interacting particle systems, with application to consensus formation or materials. Whereas the first aspect is clearly relevant for industry and offers funding opportunities from this side, the second one is more fundamental and could be relevant for EU funding schemes or even political think tanks.

## 6 Participant list

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### Organizers

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#### **Hartmann, Carsten**

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#### **Sullivan, Tim**

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**Vilmart, Gilles** - UNIGE, Geneva, Switzerland  
**Von Toussaint, Udo** - IPP - Max Planck Institute, Germany  
**Wang, Ting** - University of Delaware, United States  
**Wu, Stephen** - Institut of Statistical Mathematics, Japan  
**Zabaras, Nicholas** - University of Notre Dame, United States  
**Zhang, Wei** - Zuse Institute Berlin, Germany  
**Zygalakis, Kostas** - University of Edinburgh, United Kingdom

# Frontiers of electronic-structure theory: focus on the interface challenge

Location: DPG Meeting in Regensburg

Webpage: <https://www.cecarn.org/workshop-details/121>

Dates: Mar 31, 2019 - Apr 05, 2019

## 1 State of the art

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First-principles electronic structure calculations have become an indispensable tool in many research areas where materials surfaces or interfaces play a central role. Corresponding calculations provide insight into catalytic mechanisms, interfacial ionic and charge transport in batteries or solar cells, materials degradation through corrosion or wear, and many other highly relevant application areas. Despite this prolific use, corresponding calculations face multiple issues. Large system sizes and/or periodic boundary conditions are often imposed due to the necessity to treat the extended interfaces. Also, strong heterogeneity in geometric or electronic structure across the interface needs to be consistently described, external fields and potentials must be coupled, to name but a few of the challenges. Many applications, furthermore, require extensive numbers of these calculations to capture trends and dynamical effects, average ensembles, or provide sufficient data sets for coarse-grained simulations.

## 2 Major outcomes

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While the invited lectures had a focus on the above-described interface challenge, the symposium covered the general field of computational materials and electronic structure theory with 55 contributed talks. This was an official symposium at the DPG Spring Meeting of the Condensed Matter Section, hosted by the Surface Science Division and approximately 200 participants of the DPG meeting joined the symposium - almost 400 people of the community joined the psi-k get together on Tuesday evening. Invited Speakers and the titles of their talks at the symposium were:

- The Data Revolution in Materials Science, Through the Lens of the Materials Project, Kristin Persson (UC Berkeley, USA)
- First-Principles Approach to Model Electrochemical Reactions at the Solid-Liquid interface, Mira Todorova (MPI for Iron Research, Germany)
- Theoretical Investigations of Electrochemical CO<sub>2</sub> Reduction, Karen Chan (DTU, Denmark)
- Scaling Relations and Beyond for Kinetic Monte Carlo Models in Heterogeneous Catalysis, Mie Andersen (TU Munich, Germany)
- Addressing the Structure and Dynamics of Weakly-Bonded Interfaces, Mariana Rossi (FHI, Germany) CECAM ENDORSED THIS MEETING, BUT DID NOT PROVIDE ANY FINANCIAL SUPPORT.

## 3 Community needs

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This type of symposium should continue.

## 4 Funding N/A

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## 5 Will these developments bring societal benefits? N/A

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## 6 Participant list (Partial)

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### Organizers

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**Nørskov, Jens K.**

Technical University of Denmark, Denmark

**Reuter, Karsten**

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**Scheffler, Matthias**

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**Andersen, Mie** - Aarhus University, Denmark

**Chan, Karen** - Technical University of Denmark, Denmark

**Persson, Kristin** - Lawrence Berkeley National Laboratory, United States

**Rossi, Mariana** - Max Planck Institute for the Structure and Dynamics of Matter, Germany

**Todorova, Mira** - Max-Planck-Intitut Für Eisenforschung, Germany

# Condensed matter analogies in mechanics, optics and cold atoms

Location: CECAM-ISR

Webpage: <https://www.cecam.org/workshop-details/93>

Dates: Apr 01, 2019 - Apr 04, 2019

## 1 State of the art

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There are longstanding analogies between solid-state systems and varied physical systems. These include connections between cooperative phenomena in electronic systems and those found in elasticity, hydrodynamics, nonlinear optics, colloidal suspensions, and more. The recent interest in topological phases of matter is now seeding the search for alternative physical systems exhibiting topological phenomena. Most of these systems may be thought of as metamaterials, since they include lattices made out of some mesoscopic (or even macroscopic) repeating building blocks. There have been several recent works that demonstrated either similarities or differences between electronic or quantum systems and their analogous realizations in mechanical metamaterials, optical arrays, trapped cold atoms, artificial spin ices, and colloidal systems. However, to date there is a gap between the communities studying these different systems.

## 2 Major outcomes

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The main outcome of the workshop was the discussion and exposure across communities. People working on distant yet related fields got to learn about recent results but more importantly open questions that their colleagues are facing. Specifically, mathematical connections between topological phenomena in different systems were established. The connections between fields also enabled participants to envision potential applications of topological features in varied physical systems. Examples range from higher order topological phases that are studied in electronic and in mechanical systems, to connections between non-Hermitian quantum Hamiltonians to robotic mechanical metamaterials or odd elasticity in active fluids.

## 3 Community needs

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The workshop was very successful and we certainly think that having more such events would contribute significantly. Since we combined participants from different fields and wanted to have substantial representation to each field, the program was pretty busy, mainly with 30-minute talks. It would be beneficial in future events to have also longer, pedagogical lectures introducing not only the state of the art, but also the language and terminology used in each field.

## 4 Funding

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To our understanding, most participants have individual research grants, or research grants for small scale collaborations of a few researchers. Possibilities for larger scale collaborative grants were not discussed in the workshop.

## 5 Will these developments bring societal benefits?

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Even though the workshop was dominated by theoreticians, and discussed basic science, the fields of metamaterials and topological phases of matter have technological applications down the road. These were not discussed in the workshop, but do serve as the motivation for some of the research in these fields.

## 6 Participant list

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### Organizers

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**Huber, Sebastian**

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# Current challenges in transport, growth and dissolution at mineral-fluid interfaces

Location: CECAM-FR-RA

Webpage: <https://www.cecam.org/workshop-details/142>

Dates: Apr 03, 2019 - Apr 05, 2019

## 1 State of the art

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Mineral-water interfaces, and, specifically, carbonate minerals, are relevant in a wide range of problems in natural sciences (geophysics, geochemistry) and Society (medicine, and climate change). Carbonate-water interfaces play a key role in many physical and chemical processes: biomineralization, weathering and soil formation, carbon sequestration, scale formation, and ore mineral deposition. The investigation of these processes involves a wide range of scales, from nm to macroscopic. The advent of a new body of simulation tools, e.g. ab initio Molecular Dynamics, Monte Carlo, Lattice Boltzmann and rare event sampling techniques, has enabled the modelling of nano- and upscale processes at fluid-solid interfaces, the study of nanoparticles and complex macro-molecules and the investigation of nucleation phenomena. These techniques can bring new routes to explain the elementary steps governing nanoscale processes relevant to growth, dissolution and reactive properties of mineral surfaces. A major issue preventing the deployment of these techniques in mineral-water interfaces is the inability of specific methods to cover several length scales.

The workshop helped to define the state-of-the-art of the field and to identify the key challenges faced by different computational techniques, ab initio Density Functional Theory, Molecular Dynamics, Kinetic Monte Carlo, Lattice Boltzmann and continuum/phase field models. The participation of world-renowned experimentalists helped to discuss the suitability of these techniques to investigate the structure and dynamics of processes involving mineral interfaces.

## 2 Major outcomes

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The workshop provided a snapshot of the recent trends and novel ideas in the transport, growth and dissolution at mineral-fluid interfaces. Highlights included:

Leading researchers in the field of molecular simulations (D. Duffy and J. Gale) proposed a new consistent view of the microscopic processes involved in the growth of calcite. A new picture is emerging. Ions adsorb at preferential positions in the vicinity of atomic steps. These transition sites could play a crucial role in defining the kinetics of the growth and dissolution of calcite.

On the experimental side, important progress is done in the development of the new generation of Atomic force Microscopy in water (A. Kuehnle) allowed the observation of the multilayered structure of water in the vicinity of calcite surfaces, as well as chemical speciation. These techniques allowed us for the first time to observe directly ions at the mineral-water interface, as well as the surface defects and related structure of the liquid near the surface.

It was proposed that entropic effect play an important role in driving the kinetics of the microscopic processes taking place in the vicinity of the calcite surface.

In addition, Kinetic Monte Carlo (KMC) models (I Kurganskaya, A. Luttmge,) were discussed, showing they can reproduce crucial features of the non-equilibrium morphology of minerals (e.g., calcite, quartz, phyllosilicates) during growth and dissolution, including etch pit dynamics and the coexistence of straight/slow and rough/fast atomic steps. This later observation was confirmed by experiments discussed by J. Colombani, A. Luttmge and B. Zareepolardaniat the workshop. The new method to reproduce pitted morphologies based on the KMC parameterized Voronoi distance functions is offered (R. Rohlfs).

Motivated by the observation of biomineralization, many experimental results have pointed out the effect of additives on the growth of minerals, but also on their microstructure following growth (B. Pokroy) and dissolution (Paul Fenter), which controls their material properties (mechanical, optical, etc.).

Moving to continuum simulation methods, it was shown that Lattice Boltzmann simulation techniques can solve multi-phase fluid flow in porous media at scales that are large enough to extract directly phenomenological relations between the hydrodynamic transport and pressure gradients in the porous space (J. Mathiesen). The dissolution-precipitation processes implemented into the LB solvers showed their effectiveness in the predictions of the porosity and permeability properties of rocks and multi-grain assemblages (N. Prasianakis). The striking molecular dynamics studies (D. C. de Busturia) showed key controls of pressure induced CaCO<sub>3</sub> phase transformations. The local pressure induced phase transformations were also shown in the experiments (J. Colombani). The effects of both pressure and confinement onto the reactivity of CaCO<sub>3</sub> grains was shown experimentally (J. Dziadkowiec). The non-classical mineral nucleation pathways were discussed (B. Pokroy, J. Gale).

## 3 Community needs

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Quantitative predictions of nucleation and dissolution processes is still an outstanding objective for molecular simulations. While computational advances in the area of metadynamics have been deployed to obtain reliable surface free energies of model systems, single-component Lennard-Jones (G. Tribello), the application of such methods to calcite, involving solvent and two type of ions, has not been attempted yet.

The impact of additives on nucleation processes is still a largely unexplored topic in molecular and meso-scale simulations. The large body of experimental observations should motivate work in this area. However, this is a difficult multi-scale problem, involving not only surface processes but also phase transformations in the bulk of crystalline materials, as well as mass transport, and possibly formation of amorphous, gel-like, precursor phases.

While a large body of work in the past decade has been devoted to non-standard nucleation pathways of calcite, a better understanding of heterogeneous nucleation is crucial to understand the formation of calcite in geological and biological environments (A. Fernandez-Martinez) where effects of confinement and pressure may have a tremendous impact onto the reactive and rheological system's behavior. The upscaling of microscopic interactions between grains in granular materials is a key to understand the macroscopic properties (compaction, mechanics).

## 4 Funding

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The workshop was co-funded by the NANOHEAL Marie Curie Training Network. This network has contributed significantly to the advances in the understanding of the non-equilibrium processes involved in mineral growth, dissolution, from the nano-scale to the continuum scales. A novel training network based on the existing network has been submitted to the European Research Council. The success of this project is important to keep Europe at the leading edge of research in the field, and to maintain and build synergies in Europe.

Despite many applications of mineral growth and dissolution in industry, there is need for funding long-term fundamental research to keep the European community active. Also, in-out from the European Council is essential to sustain research in this interdisciplinary topic.

## 5 Will these developments bring societal benefits?

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The primary aim of the Workshop was bringing together scientists working on computational and experimental approaches covering multiscale aspects of mineral surface reactions: growth, nucleation inhibition and chemical and physical processes under nano-confinement conditions, where the reactive system size is limited spatially. The workshops stimulated discussions between scientists working across scales: nano, micron and mm-size pores, and from nanoparticles to macroscopic mineral grains. Carbonate minerals, e.g. calcite, represented a central aspect of the discussions and contributions.

Short-term or long-term societal benefits should emerge from the understanding of transport, growth and dissolution at mineral-fluid interfaces. Some examples include:

- carbon sequestration as well as sequestration and storage of toxic and radioactive waste
- drug delivery (calcite with additives allows for slow release of drugs due to its slow dissolution dynamics)
- bone/teeth replacement (challenges are to produce replacement materials with suitable material properties and compatibility to animal physiological conditions).
- cell and other material cryopreservation,
- concrete: (i) production of green sustainable concrete; (ii) improvement of the properties of concrete from a better control of the dissolution of various minerals, and of the micro/nano-structure assisted by additives. (iii) extrapolation of concrete properties to very long times for the sequestration of radioactive waste.

## 6 Participant list

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# Perspectives and challenges in modeling of small angle scattering data in soft matter and life sciences

Location: Planned location: Institut Laue-Langevin (in collaboration with CECAM-FR-RA)

Webpage: <https://www.cecarn.org/workshop-details/88>

Dates: Apr 08, 2019 - Apr 12, 2019

## 1 State of the art

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As the field of modeling in Soft Matter and Life Sciences is growing tremendously, the gap between the expertise of dedicated research groups and the capabilities of their software on one side, and the knowledge of Small Angle Scattering users on the other side, is becoming abyssal, with fortunately a (slowly) increasing number of exceptions. This workshop aims at bridging these two worlds. Small Angle Scattering (SAS) is a non-destructive technique that measures heterogeneities in the bulk with no specific sample preparation. The probed volume of sample varies from 0.1 to 1000 mm<sup>3</sup>, i.e. the data obtained are statistically highly relevant. One SAS spectrum contains all information in the reciprocal space corresponding to the ~1-1000 nm direct space, and SAS can be acquired on samples under external or internal stress. It is therefore completely different, and complementary of cryo-EM techniques. Today, the majority of users of SAS instruments in Life Sciences rely on the EMBL software package ATSAS, while a number of other solutions developed also focus essentially on biomacromolecules (see the online project SASSIE or the recent Pepsi-SAXS). These user-friendly tools are largely limited to simple situations of infinitely dilute solutions, where either the relevant unit is small and contains a structurally well-defined unique material, allowing to perform MD simulation. In parallel, the majority of users in Soft Matter rely on analyses with analytical equations derived from relatively simple geometric considerations. Because well-established 'traditional' software does not account for more complex situations, both communities are actually limiting their research to what the known approaches can deal with.

## 2 Major outcomes

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- Discussions between theory, simulation and SAS experimentalists communities: Degree of confidence in data, modelling and comparison techniques.
- Presentation and discussion of different software packages. Advantages and specificities.
- Possibility for the users to try different packages to choose the ones that are adapted for specific scientific questions the best.
- Agreed on the community benchmarks - both on experimental systems measured on multiple instruments world-wide, and computational packages for the analysis of experimental results. Preliminary agreement on the benchmark publication.
- Agreed on writing a multisoftware tutorial to guide the users what and how to use for a particular system in hand.

- Agreed on further workshops - this one will be continued in the current format, and another one will be organised for integrative structural biology.
- One of the main open questions - how to properly integrate experimental knowledge into modelling pipelines. One of the best suggestions was to use as much as possible the maximum likelihood approach and the Bayesian statistics.
- Multiscale modelling is still a challenge - the community wonders if it will be possible in 15 years.
- We agreed that raw data  $I(q)$  should be used in comparison of simulations with the experiments.
- The question of estimation of actual information content has been raised.
- Current software techniques cannot integrate multiple experimental data (SAXS and SANS), mix analytical models with atomistic representations (except the WillItFit approach), and rigorously model effects of solvation.
- We agreed on sabbaticals spent in different labs and more student exchange.
- More general models for the solvation shell are required.

## 3 Community needs

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1. Software developers need a database with high-quality curated small-angle scattering data. The current 2 databases (SASBDB and BioISIS) are very limited.
2. A community analysis has to be performed that demonstrates how to properly model and compare atomistic systems with experimental data.
3. We need access to raw detector data before reduction - this will be useful to the machine learning community.
4. More tutorials from multiple developers and multiple experimental teams are needed.
5. Strength of the network between experimentalists and modellers.
6. There is a clear need for more workshops of this style joining experimentalists, software developers and theoreticians.
7. The outcomes of these workshops should be shared with the specialised master programs in structural bioinformatics and structural biology.
8. HPC is not the bottleneck in the community.
9. More efforts must be put into the development of multiscale modelling and multitechnique data integration. Close mimicking the experimental conditions is also needed.
10. Better software and more benchmarks for neutron experiments are needed.

## 4 Funding

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National and international experimental facilities (e.g. Institut Laue-Langevin, ESRF The European Synchrotron) are typical funding sources. ERC and national funding agencies also contribute. Joint research proposal (including joint PhD proposals) have been discussed. We have also discussed upcoming workshops and tutorials.

## 5 Will these developments bring societal benefits?

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Small-angle experiments are getting popular in chemical and pharmaceutical industry. This involves multiple axes: - the new generation of drugs is based on peptide interactions with proteins or membranes. However, peptides are very flexible systems not well suited to be studied with classical techniques such as X-ray crystallography. Therefore, there is strong need for characterisation of their complex behaviour in solution using additional techniques that largely include small-angle scattering (both SAXS and SANS), and also specific software and algorithms. - another current academic and industrial research topic is intrinsically disordered proteins. Again, these cannot be studied using classical techniques and much effort has been invested to their study using SAXS and SANS. However, much more progress is required in this field, especially on the computational side. This is due to the gigantic conformational space probed by these macromolecules at physiological conditions. - the future industrial needs will include softmatter systems measured and simulated using multiscale approaches. We should also add that industry uses the same software techniques as the academic users. Regarding the funding opportunities - there multiple options including the InnovaXN program between ESRF, ILL, and EU, national grants (e.g. French government fund for innovation clusters), and more.

## 6 Participant list

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### Organizers

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# Microscopic simulations: forecasting the next two decades

Location: University Paul Sabatier, Toulouse, France

Webpage: <https://www.cecam.org/workshop-details/107>

Dates: Apr 24, 2019 - Apr 26, 2019

## 1 State of the art

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Theoretical, methodological and algorithmic developments require long term research investments. It is well accepted that developing a simulation code needs from 10 to 20 years of effort. This is also observed in the field of microscopic simulations where systems like finite size nanodrops, polymers/bio-polymers and bulk/vapor interfaces, are simulated at the atomic or molecular scale. For instance, the molecular modeling code CHARMM and the quantum chemistry package of programs GAUSSIAN were first developed in the seventies and they are both still among the most popular codes used today. The popular codes NAMD, GROMACS and LAMMPS or the quantum molecular code CPMD are also still developed after more than 20 years. Regarding methods, we have obviously to mention here the never ending race to refine force fields and the case of polarizable force fields that were proposed and discussed as soon as the seventies and that started to be intensively used during the last decade. Developing a theoretical method and writing a simulation code may be considered as two distinct standalone activities. Nevertheless, codes can be developed to allow the use of new theoretical methods that still needs further improvements. We may also note the outstanding increase of the available computational resources. In about 15 years, the power of a typical computing system available in national centers has increased from dozens of Tflops to now hundreds of Pflops, with Exaflops systems on the horizon. While the computing system panorama was stable over the last decade, dominated by standard and almost monolithic INTEL CPU-based architectures, we are now facing an important evolution. First more than 56% of the computational power available in the fastest supercomputing systems results from GPU units and new actors are emerging proposing new architectures based on ARM computational units and even computing systems specifically devoted to molecular dynamics (the ANTON machine, developed by the D.E. Shaw Research Laboratory). We have thus to be aware of the forthcoming new generation of computing systems to propose not only interesting but also efficient new theoretical methods and algorithms. On the experimental side, the main features and capacities of experimental apparatus also evolve rapidly. Decoding a full human genome can be performed today at the week scale by a single team, while years of efforts were needed by a large international network of research centers only 20 years ago. This means that bio informatics developments today have to account for the amazing amount of storage capacity needed to store these genetic data and for all the problems tied to analyzing these data, problems that were far from being critical 20 years ago. Microscopic simulations are considered as a promising alternative route not only to interpret experiments but also to complement them (for instance to “feed” data banks about ion thermodynamics properties in liquid phase). As discussed above the development of simulation codes and theoretical methods cannot be considered as standalone activities. They have to be driven to complement experiments, to match potential new needs and to be well suited to be used on the forthcoming high-performance computing systems in order to reach the highest level of efficiency when performing simulations. It is thus pivotal to anticipate as soon as possible (and as far in the future as possible) what will be new potential needs and the computational state of the art in the forthcoming decades, in order to initiate the development of new generation of codes and of theoretical methods that will be used by large communities, from basic research to industry.

The objective is obviously not to cover all the possible research fields and not to identify all the possible new methods and codes to develop in the forthcoming years. We will focus the workshop on topics related mainly on simulations in liquid phase and at liquid/vapor interface. Nevertheless, our aim is to provide a clear overview of the different however inherent parameters that have to be accounted for by young theoreticians who want to start the development of new theoretical methods and codes with the aim that these methods and codes will be used by the largest as possible communities, from basic research to industry and regardless of their research field. Moreover, the aim is also to show these key parameters to be the same regardless the code application fields.

## 2 Major outcomes

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The aim of the workshop was to stimulate discussions among actors from different fields (like mathematicians, computational science, biology and atmospheric/astrophysical chemistry experts as well as experimentalists) to identify the major directions and problems that we will have to face in the forthcoming decades in the microscopic simulation research field. As recalled by a participant, Niels Bohr stated “It’s difficult to make predictions, especially about the future”. However, the discussions we had during the workshop allowed us to at least identify several important issues/problems that microscopic simulations are facing or will have to face in the forthcoming decades:

1. The most important issue raised up by several speakers is the reliability of the interatomic potentials in describing accurately microscopic interactions, the so-called “force fields”. Despite decades of efforts, the available force fields, in particular pairwise ones, are still considered as not accurate enough to model biological systems, ice at low temperatures and water/alcohol mixtures for instance. Moreover, new needs for accurate force fields have emerged in the atmospheric and astrophysical chemistry modeling where experiments regarding complex and finite size systems are more and more difficult to interpret. However recent progress in quantum ab initio methods suggests that it will be easier electronic structure methods at the complete basis set limit. Instead of considering exclusively experimental data, quantum-based data offer an interesting alternative way for assigning accurate and transferable force-field parameters. We may also note that an original 'reactive' force field approach was presented during the workshop, paving the road towards efficiently investigating chemical reactions active solvents such as water and alcohols.
2. Besides force-field accuracy, another important barrier to the effective use of microscopic simulations (in particular for industrial applications) is the necessity to generate simulations that are large enough and long enough to provide statistically relevant results. For instance, several speakers underlined the need of performing molecular dynamics simulations of large systems at the 100 ns scale and even longer, preferably in a few days. However, available microscopic simulation codes are not adapted to modern massively parallel CPU-based computational systems. From the discussions with computational science experts, to take advantage of the modern computational infrastructures and the hardware evolution anticipated in national/European computational centers (GPU, ARM etc...), over the next five years, a massive action involving a rewriting of the actual simulation codes has to be achieved. Fortunately, the use of proper analysis tools can greatly reduce the cost of rewriting and in particular provide accurate diagnostics in terms of performance gains. This is a major conundrum questioning the pertinence of new developments aiming at only increasing further and further the efficiency of the available codes (and the methods they rely on), in particular by the usual small teams working in the microscopic simulation field. Moreover, the simulation length issue is also a major limitation

preventing sophisticated force fields to come to maturity. A proper balance between accuracy and efficiency is critical.

3. On the positive side, new theoretical methods, developed in particular by mathematicians are shown to reduce by several orders of magnitude the simulation length needed to get statistically relevant results (like the Adaptive Multilevel Splitting algorithm to speed up the sampling of reactive trajectories, for instance). That suggests to go on favoring discussions between microscopic simulation experts (in particular physicists) and mathematicians. That should lead to new numerical methods able to tackle the simulation length issue. However, attention has been paid at the algorithmic level to favor regular data structures and regular flow of operations. Taking such constraints into account from the start will allow a better exploitation of the upcoming HPC processors. Hence, discussions between physicists, mathematicians and computational experts should also be favored.
4. Regarding the present numerical revolution arising from machine learning and AI techniques and from new actors in the microscopic simulation field (like Google DeepMind whose teams recently participated in an international protein folding blind test competition), our discussions highlighted that for these techniques to improve the predictive capacity of protein folding tools, for instance, large sets of training data are required. The current availability of such large data sets for microscopic simulation applications (to improve force fields for instance) is a priori poor, not to say nonexistent. However, promising applications of machine learning techniques to analyze molecular dynamics simulation trajectories have been presented. Regarding AI techniques, some specific problems, like reproducibility issues or inadaptability to propose new models, have been also discussed. These considerations clearly suggest to “thinking differently” when trying to employ AI in the context of microscopic simulations. An interesting application of AI noted by some participants would be to improve the user experience and interactivity with molecular simulation tools, for instance to facilitate parameterization.
5. A major challenge also mentioned, that requires further discussions, concerns the training of the PhD students who should develop skills in chemical, biological and physical understanding but also integrate all the above-mentioned concepts, a breakthrough compared to the actual situation. Moreover, these students should be trained to adopt good practices when writing codes, such as a systematic use of the git repository. Our discussions also pointed out the necessity of providing source codes to journal editors when publishing new methods (or new results based on “home” codes), even if that means extra efforts for the smaller research teams working on the developments and/or implementation of new theoretical methods.
6. Last but not the least, the discussions during our workshop clearly show the need to noticeably increase the visibility of the teams working in the microscopic simulation field, (a) to favor their participation to European R&D programs like EURATOM and (b) to favor new collaborations between academic and industrial teams.

We may note here that some industrial scientists emphasized their difficulty in identifying academic experts able to provide pertinent solutions (method/model/code etc.) to their problems. This lack of visibility also prevents those who are actively involved in improving simulations from adequately defending their point of view during discussions about future European computational infrastructure.

## 3 Community needs

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Regarding the community needs, several participants expressed their willing to participate to a network involving mathematicians, computational scientists, experimentalists and industrials. To our opinion, such a network labelled by the CECAM should be a major tool to go on developing the field of microscopic simulations and to strengthen the visibility of simulation experts. Moreover, such a network may also favor the actors of the microscopic simulation field to defend their point of view during discussions about future European computational infrastructure, for instance. From the discussions with the participants, we have already decided to organize a workshop on the same topic within two of three years.

## 4 Funding

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Typical funding channels are ERC grants or national grants (like ANR in France). However, we identified new funding channels from European Research Programs like EURATOM and its forthcoming calls for the period 2020-2025, for instance. From the workshop discussions, at least five meetings to start new collaborations between academic research teams and/or between academic researchers and industrials have already been planned. These meetings will in particular lead to new funding channels (mainly from industry) to promote molecular simulations.

## 5 Will these developments bring societal benefits?

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As discussed in the above section 4, some industrials propose to collaborate with academic research teams and to fund their research activity. Their aim is a priori to adapt available methods or the develop new ones to meet their own needs. It has to be noted that industrials clearly expressed their willing to go on with such workshops allowing both to create new networks as well as to discuss about their needs.

## 6 Participant list

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**Lynne, Soderholm** - Argonne National Laboratory, United States

**Quignot, Chloé** - CEA, France

**Sanchez Marcos, Enrique** - University of Sevilla, Spain

**Severo Pereira Gomes, André** - CNRS, France

**Steffenel, Luiz Angelo** - Université de Reims Champagne Ardenne, France

**Stoltz, Gabriel** - Ecole des Ponts, France

**Toubin, Céline** - PhLAM Laboratory, France

**Traoré, Seydou** - Neoxia, France

**William, Jalby** - University of Versailles-St Quentin, France

# Network analysis to elucidate natural system dynamics, diversity and performance

Location: CECAM-FR-RA

Webpage: <https://www.cecarn.org/workshop-details/115>

Dates: May 02, 2019 - May 04, 2019

## 1 State of the art

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Omics-Data has brought into light the fact that nothing is unique in living systems. Whatever the scale, the building blocks of life are based on diversity and whatever collective acts they orchestrate, they use many alternative approaches. The diversity lies at the level of individual elements and at the level of their connectivity making network-based modeling approaches relevant to investigate the role of diversity in the properties of living systems.

- The first day of the CECAM focused on modeling experimental data and molecular dynamics simulations using network-based models to probe molecular and protein dynamics and structures. Small enzymes to big protein oligomers were studied using mass spectrometry or time-resolved infra-red spectroscopy for example.
- The second day demonstrated the wide range of applications of the network approach to biological systems spanning from the brain (cm) to the genes (nm) passing by the cellular level (micron). In addition, several PhD students and young scientists were invited to present their work.
- The third day focused on deeper technical issues using protein structure networks. Again the features of large-scale propagation and hierarchical organization were shown. Essentially, the multi-scale aspect of protein dynamics and the propagation of motions/ energy/ geometrical changes across the system were investigated and the systems displayed non-linear properties. Interestingly, DNA expert also presented results indicating the potential application of networks to explore DNA dynamics as well. All the systems showed hierarchical architecture and multi-scale dynamics. Network approaches have the advantages to infer dynamics features even from static structural data because the network model sits between the chemical composition of the molecules and their function.

## 2 Major outcomes

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The workshop main outcome was to gather a very multidisciplinary group, which discussed several themes in the biology field, on both experimental and theoretical grounds. The network approach was demonstrated to provide original insight in many biological systems. The talks and discussions also provided evidences of the network paradigm as a bridge between theory and experiments, so to model real systems from the many data available in biology. What called our attention was the specific properties of natural systems in terms of complex systems behaviour - biological networks are very close to the percolation threshold, that means they are very sparse yet still connected and working. As a result, they allow long-range transport through systems of biomolecules - biological networks are designed to be sustainable, i.e. to

be robust and adaptable to perturbations. While mutation may change details of the network, rearrangement of the network as a result of perturbation still maintains long-range channels and signaling. -protein dynamics and allosteric regulation was one of several topics addressed at the meeting. Networks connecting and providing signaling between ligand binding sites have been identified by a number of computational tools, as well as ligand binding sites and binding sites for heterotropic effectors. -biological networks and their characteristics were explored by a number of computational tools discussed at the meeting. New research directions were identified, in particular tools applied to specific problems by one participant will be combined with tools adopted by other participants to further examine properties of networks and their role in function in a wider range of systems. Several collaborations were settled at the workshop. Luisa di Paola joined Claire Lesieur's lab for one month. Lorenza Pacini visited Matteo Degiacomi's lab in the UK for one month. David Leitner is visiting the lab of Takahisa Yamato at Nagoya University for almost four months (Fall 2019), and the lab of Tamiki Komatsuzaki for a week in October 2019, two other participants. During those visits the workshop participants plan on following up on ideas discussed at the workshop on energy transport networks in proteins and protein complexes. Main limitations are the infrastructural resources for more burdening computational applications (molecular dynamics of large systems) and the necessity to build up and feed a multidisciplinary network of researchers in different fields, to share perspectives and methodologies. The contamination between different scientific fields on a common ground (network approach) has already proved to lead to interesting results and innovative methods.

## 3 Community needs

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The computational needs may be classified in:

- infrastructural: the need of more computational resources for more burdening computational activities (molecular dynamics of large macromolecules and complexes)
- the accessibility to share web portals for shared computational applications; Additionally, more general needs include:
- widening the community to other researchers in applied fields (pharmaceutical applications, ecosystem webs, etc...) and to the complex systems community.

The first will allow us to see how general the natural system network properties are and whether these properties can be used as a source of inspiration and innovation. Second, the natural properties call on new theoretical network models and simulation to track the cause of their emergence.

In order to meet these needs, we propose the following strategies:

- To keep the community alive and improve visibility we need communication tools (community webpage, newsletter, blog);
- Likewise, events can improve the community visibility and consolidate the common activities.

It was discussed the possibility to have an event every two years, alternatively a CECAM workshop (specialized conference) and a satellite workshop at the Conferences on Complex Systems for example to reach other communities.



## 4 Funding

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The CECAM workshop received additional fund support from the University of Nevada (Reno, USA), Institute of Complex System of Lyon (IXXI-ENS Lyon), GDR BIM (Bioinformatics Molecular). We further discussed the possibility of joint applications (e.g. HFSP, European applications), including funds for conference/workshop organization.

## 5 Will these developments bring societal benefits?

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At present, the society is facing complex and urgent issues related to the climate degradation and its consequences on human health and biodiversity. We, as scientists in network modeling are in good position to address these issues and propose appropriate solutions by understanding the connectivity between multiple individual elements and the large collective actions born from them. This knowledge is necessary to diagnose and correct errors related to genetic mutations (genetic diseases, health) or unbalanced ecosystems. Understanding the details of natural networks also provides more sustainable design solutions to men-made systems. This is one reason to broaden the approach to other systems and work with other communities.

## 6 Participant list

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### Organizers

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**Di Paola, Luisa**

University Campus Biomedico Rome, Italy

**Giuliani, Alessandro**

“Sapienza” University of Rome, Italy

**Leitner, David**

University of Nevada, Reno, United States

**Lesieur, Claire**

CNRS, France

**Papaleo, Elena**

Danish Cancer Society Research Center - Denmark, Denmark

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**Caselle, Michele** - Università di Torino, Italy

**Degiacomi, Matteo** - Durham University, United Kingdom

**Fanelli, Francesca** - UNIMORE, Italy

**Fraternali, Franca** - King's College London, United Kingdom

**Junier, Ivan** - Univ. Grenoble Alpes, France

**Knoblauch, Kenneth** - SBRI, France

**Komatsuzaki, Tamiki** - Hokkaido University, Japan

**Naganathan, Athi N.** - Indian Institute of Technology Madras, Chennai, British Indian Ocean Territory

**Rivalta, Ivan** - Ecole Normale Supérieure Lyon, CNRS, France

**Siegel, Anne** - RISA - CNRS, France

**Stock, Gerhard** - University of Freiburg, Germany

**Tiwary, Pratyush** - University of Maryland, United States

**Vuillon, Laurent** - Université de Savoie, France

**Wales, David** - Cambridge University, United Kingdom

**Yamato, Takahisa** - Nagoya University, Japan

# Young researcher's workshop on machine learning for materials science

Location: CECAM-FI

Webpage: <https://www.cecarn.org/workshop-details/91>

Dates: May 06, 2019 - May 10, 2019

## 1 State of the art

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The influx of machine learning (ML) algorithms from computer science into computational materials science (MS) have led to developments of novel computational methodologies and opened up novel routes to addressing outstanding problems. Seminal works have combined electronic structure simulations with ML algorithms in global structure search, to predict stabilities of molecules and solids, or target materials with particular functional properties. Much has become known about the performance and versatility of ML techniques such as kernel ridge and Gaussian process regression, neural networks, genetic algorithms and Bayesian optimization, with recent advances too numerous to mention. Materials descriptors that facilitate ML for both molecules and solids are growing ever more sophisticated and have dramatically improved the accuracy of ML predictions. Young researchers are however not necessarily equipped with the right background to promptly and fruitfully use them in their research. Our workshop was aimed at nurturing the ground for the next generation of scientists, which will be able to proactively and efficiently exploit data-driven techniques in the field of materials modelling. To this end, we brought together young researchers, experts from computer science, and world-wide renowned researchers applying ML in computational physics or chemistry. Didactic introductory lectures as well as hands on tutorials introduced the researchers to a broad range of machine learning techniques and algorithms. Computer scientists presented additional algorithms and techniques that could help overcome the shortcomings of current methodologies and develop approaches beyond the state-of-the-art. Timetabled discussion sessions were organised to promote cross-talk and nurture scientific advancements; these focused on key issues such as: generalising materials descriptors, inverse learning problems and generative models; latent space and reinforcement learning approaches; quantifying uncertainty in ML predictions.

## 2 Major outcomes

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A major outcome of the ML4MS 2019 workshop was the training of young scientists during the first two days of the event. Out of 149 participants, 115 were students (77%) who benefitted from the 80min introductory lectures on key aspects of machine learning methodology currently in use in materials science research. There was also much interest in the one-day computational tutorial, which was sponsored by the CSC - IT Centre for Science, the Finnish national supercomputer centre. In addition to the 45 participants accommodated in the CSC classrooms, the tutorial was live-streamed to the conference room, where the rest of the students participated in the tutorial exercises on their own laptops. In total, 91 students completed the exercises, making this the largest CSC training event to date. The feedback to the training events has been particularly positive: 75% participants said they would employ new knowledge in their own work (ML4MS feedback survey). In the latter part of the workshop programme, the most interesting ideas came during the panel discussions sessions with the

invited speakers. Experts from computer science in particular brought novel insight to the panels. During the two sessions, the Presemo online tool was used to collect questions from the participants: they posted new questions, and voted for the existing questions they thought were relevant.

- There was a debate on the importance of interpretability of ML models in MS. Experts from computer science community pointed out that reliability, and therefore estimates of uncertainty, ended up being more important than interpretability in other research fields.
- Feature engineering in material descriptors was identified as an important way of encoding subject matter into ML investigations, instead of relying on very big datasets.
- It was highlighted that latent spaces involve compressed information about the MS problem, and suggested that sampling be performed in latent spaces, instead of high-dimensional physical ones.
- Sharing computer codes was identified as good practice in the advancement of this research field, but also a danger for inexperienced scientists. When sharing code, it is important to highlight the regime of validity of the code/technique
- It was discussed why learning with graphs and strings is very successful, compared to structure (3D) based descriptors. On one hand, generative models for sequences are historically very well developed. On the other hand, material design targets can be also achieved with graph networks which describe a 3D manifold, without the need for a full 3D descriptor.

## 3 Community needs

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Machine learning applications in material science constitute a relatively young research field, but one that is rapidly expanding. We estimated 50-80 participants but received 200 workshop registrations, 3 times more than anticipated. The enormous response we had from young researchers to our introductory lectures and hands-on exercises suggests there is a need for further school and training events. At present, most schools in computational physics and chemistry focus on simulation methods with a peripheral coverage of machine learning topics, so further events that train exclusively and widely in machine learning are needed. Likewise, the growing interest in machine learning in materials science is reflected by the content of CECAM workshops. In 2019, four different events featured the topic “machine learning” in the title, with the author aware of at least 4 ML-related Psi-K ones. The influx of new methodology is clearly revitalising computational research in our community. More formal and structured events would help to bring the grassroots efforts of the community together for a more systematic advancement of the field. Our workshop already identified the need for code sharing, creating new resources and infrastructures, perhaps facilitated by the relevant European Centres of Excellence. To bring together the wider community of researchers, and keep facilitating the education of young talent, we argue that this workshop should become a series. Reaching out to the computer science community was extremely helpful, and future events should include experimental science as well.

## 4 Funding

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While some of the work presented at the workshop was funded by the NSF, Horizon 2020, NOMAD and MARVEL centres, many studies reflected grassroots efforts to take up machine learning methodology. These will feature in new grant proposals to national and international

funding schemes, mostly European since 90% of the participants work in Europe. 30% of participants received feedback on their own work (ML4MS survey), which means scientifically stronger proposals. In fact, 51% participants obtained useful new contacts from the event, and 8% said they would start collaborating with persons they met (ML4MS survey, 53 responses). The possibility of joint research proposals was clearly discussed during this meeting, albeit privately.

## 5 Will these developments bring societal benefits?

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At this workshop, not only did we bring the computer science and materials science disciplines together, we reached out to the industrial companies. Industry talks at scientific workshops are rare, but we wished to inform our community about the needs of the industry and ways this methodology is employed outside of academia. We hosted IBM Finland and IBM Zurich, who spoke about how ML is used both in materials research and AI platform services. NVIDIA described how ML is employed for autonomous driving, and Curious AI described their AI consultancy services. It was illuminating for our audience to understand the role ML methodology in industrial projects and how it can achieve societal benefit. The activities of IBM Zurich were particularly related to the topics of ML4MS. Similar developments in future ML4MS events have the potential to influence industrial research and disseminate into other industrial services too. In future events, we would consider expanding the participation of the industrial partners to an entire session, and organising a panel discussion to facilitate further interaction with participants.

## 6 Participant list N/A

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### Organizers

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**Foster, Foster**

Aalto University, Finland

**Glielmo, Aldo**

King's College London, United Kingdom

**Rinke, Patrick**

Aalto University, Helsinki, Finland

**Rossi, Kevin**

King's College London, United Kingdom

**Todorovic, Milica**

Department of Applied Physics, Aalto University, Finland

**Zeni, Claudio**

King's College London, United Kingdom

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# Challenges in large scale biomolecular simulations 2019: bridging theory and experiments

Location: Institut d'Études Scientifiques de Cargèse, France

Webpage: <https://www.cecam.org/workshop-details/147>

Dates: May 13, 2019 - May 17, 2019

## 1 State of the art

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The essential challenge posed by human health requires the understanding of the cell's machinery at a molecular level. The interplay between proteins, DNA and RNA is key for vital functions such as DNA transcription, translation and epigenetics. To understand these processes many experimental techniques are put in action, spanning a very wide range in terms of spatial resolution, temporal resolution and level of detail with which they can observe the macromolecules. As for all branches of science, theoretical (modeling) and experimental approaches have been developed over the years to study these systems, and, with no surprise, the most successful strategies are those for which the two approaches come together to give a full picture of the system. Indeed, because of the diversity and complementarity of the experimental techniques, molecular modeling becomes a necessary tool to decode experimental data, bridging different sources of information and building a coherent structural model compatible with experiments. From the early days physical modeling played an important role in the two principal experimental method for high-resolution: X-Ray crystallography, requiring an initial model for phasing, and nuclear magnetic resonance (NMR), requiring a multi-dimensional minimization process on a model to infer possible structures. With the current capabilities of molecular simulations (MD) the contribution of modeling can now go much further than structure refinement. In recent years physical modeling and simulation techniques started to be coupled to experimental data in order to obtain an understanding of the biomolecular systems from an atomistic description all the way up to the meso-scale.

## 2 Major outcomes

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The workshop had the aim of bringing together modelers and experimentalists. Out of the 40 speakers 8 of them were experimentalists who, as asked by the organisers, focused their presentations on the method used for their investigation as well as on the specific problem of their research. This allowed to have a view of the centers of interest of experimentalist and to have more insights on where modeling can come to their help. The other presentations were an equal mix of method development and applications to biological systems. Among the topic covered there were aspects related to sampling techniques and force field development, that are still a key point of our work far from being resolved, aspects related specifically on how to integrate experimental data and simulations, and applications to large systems such as proteins in crowded environments, cellular membranes, capsids, amyloid aggregates, ... One of the main aspects that emerged from the workshop is that we now have the tools and the computer power to tackle heterogeneous systems, including several cellular components.

However, to guide these studies, the integration of experimental data is essential both as a post-simulation comparison, and as a bias/ guide toward solutions compatible with experiments. In this respect more work is still needed to develop methods to integrate experimental data, both on theoretical grounds and on their applications to commonly used simulations software.

## 3 Community needs

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During the meeting we extensively discussed about the need to consolidate our community and to give it a permanent status, giving it visibility at the international level and access to some funding. The main financial need is to maintain and expand collaborations within the network and to have meetings on a regular basis. The methods we develop are to a great extent complementary. To tackle the largest challenges in modeling we need to bring these methods together. Therefore, a tight network is needed to develop methods on a larger-scale. The development of this network will allow us in the future to work even more closely with experimentalists. A critical point raised is the relationship with experimentalists that are still often very skeptical of our methods. We believe that we need to show them on some real problems what we can achieve and how our work can be useful to them. Along these lines we agreed to have a meeting in the future where we work collectively in solving a problem brought to us by an experimentalist. We proposed to attempt this problem-solving workshop either in 2 or in 4 years, in a European follow-up meeting of the series. Given the broad scope of the series of meetings that have alternated between the US and Europe since 2015 and the reasons just explained, a series of CECAM workshops (every other year) on the topic of large-scale biomolecular simulations would be greatly useful.

## 4 Funding

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This year we had two sources of funding beside CECAM based in France. We had 2200 euros from the Corsica Region, related to the location of the meeting, and 1500 euros from a French research group on bioinformatics (GDR BIM). The overall amount we had at our disposal was of 9700 euros, which was enough to cover fixed expenses, but not enough to cover personal expenses of participants. Ideally, we would like to have some funding to give contributions to participants, especially for those coming from outside Europe, given the higher traveling costs, and for the younger scientists, including young experimentalist that we wish to sensitize to modeling. On a small scale, to try to satisfy the financial needs of possible future meetings, we decided to create share resources between all participants to record all possible local funding opportunities. On a more global scale, we are looking into creating a network at the European level, applying for example to a COST action.

## 5 Will these developments bring societal benefits?

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The societal benefits of our research are manifold, spanning from technological application in polymer science to pharmaceutical applications in drug design. Most of our work focuses on the understanding of biomolecules and their role in associations to disease. Alzheimer and cancer are two examples which have been extensively discussed in the meeting. The application of our methods can help understand experiments and obtain an atomistic view on the action of a disease and therefore allow efficient drug design. The work of some of the organisers is already inscribed along this line, given their belonging to a Pharmaceutical Science department. The studies more focused on material and polymer science have an impact on the development of green technologies and sustainability, for example dealing with water purification.

## 6 Participant list N/A

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### Organizers

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**Chebaro, Yasmine**

CNRS, Institut de Génétique et de Biologie Moléculaire et Cellulaire, Strasbourg, France

**Coluzza, Ivan**

Center for Cooperative Research In Biomaterials, Spain

**Frezza, Elisa**

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**Nicolas, Leulliot**

Université Paris Descartes, France

**Pascuali, Samuela**

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**Schlick, Tamar**

New York University, United States

**Sterpone, Fabio**

IBPC and Université de Paris, France

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# Biomolecular mechanisms at functionalized solid surfaces

Location: CECAM-FR-MOSER

Webpage: <https://www.cecarn.org/workshop-details/139>

Dates: May 14, 2019 - May 17, 2019

## 1 State of the art

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The interaction of biomolecular systems with solid surface is of crucial importance in the field of biomaterials, since it plays a key role in numerous applications, such as tissue engineering and regenerative medicine (where we need to know the cellular response to implanted materials), the optimization of surfaces for biosensors, the development of bioactive nanoparticles, biocatalysis or bioanalytical systems. This phenomenon has now been under scrutiny for several decades, but reaching a detailed understanding of the molecular mechanisms associated with biomolecular adsorption on functionalized surfaces is far from being achieved. For over ten years, molecular simulation approaches have been developed to address these issues and represent a promising tool for the biomaterials field. Molecular simulation approaches can roughly be divided in three classes. Numerous works have been made using quantum mechanical (QM) computations at the solid interface but, due to inherent size limits of QM approaches, these works cannot encompass the complexity of biological systems. Therefore, researchers also resort to all-atom empirical force-fields and coarse-grain methods, which grant a compromise in accuracy in the description of the modeled system and thus feature decreased computational costs. The workshop gathered roughly 35 participants, including students and experienced researchers, mostly theoreticians. However, one session was devoted to the experimental approaches developed to investigate the Bio/Solid interface and several talks highlighted close collaborations between experimentalists and theoreticians in the field.

## 2 Major outcomes

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The following key issues for the development of modeling approaches were addressed during the talks and the final round table:

- The need for empirical force-field parametrization. In particular, the force-field transferability is an important question, since most FF parameters for biomolecular systems such as proteins and nucleic acid were developed for molecules in solution and might not be suitable for the same molecules in a different environment (such as the vicinity of a solid surface). In addition, one also has to develop specific FF parameters for the numerous possible solid surfaces, which can be metallic, silica or carbon-based, and bear various functionalizing chemical groups.
- Solvation effects, which can be modeled using explicit or implicit representation, are essential, since solvent molecules are active components of the system, and likely to play a role in the molecular processes taking place on the surface. In the case of an explicit representation, the question whether the surrounding solvent molecules are in equilibrium is an important one.

- Sampling is another critical aspect to take into account. In fact, events taking place on bio/solid interfaces happen on long time-scales, and their investigation requires the modeling of large systems with a rough energy surface, that are difficult to explore thoroughly for an accessible computational cost. In that perspective, multi-scale modeling approaches offer new perspectives for exploring a large range of time and length scales. However, hybrid multi-scale approaches, such as QM/MM, require to define precisely the part of the systems that will be described with a high resolution (quantum approach). This could be done with surfaces presenting well defined binding sites, but could be more problematic for an homogeneous infinite surface. Potential inspiration could be found in approaches that are currently developed to model polymers on solid surfaces.
- Another possibility to improve sampling is to develop integrative approaches that take into account data from various experimental approaches (NMR spectroscopy, PMIRAs...)

## 3 Community needs

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- Simulations including biomolecules and solid surfaces are still non-standard in the molecular modeling field and the scientific community needs the development of new force fields to describe the energy between the interacting partners.
- In particular the reactivity of the solid surface, and the possibility for the adsorbed biomolecule to form covalent interaction with it, needs to be taken into account.
- In addition, most nanoparticles surfaces are often functionalised (to avoid their aggregation), for example with SAMs, which extends further the range of components whose interaction with biomolecules need to be parametrized.
- Finally, the discussion during the roundtable highlighted the lack of online resources (such as tutorials) for researchers in the field.

## 4 Funding N/A

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## 5 Will these developments bring societal benefits?

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Understanding the interaction between solid surfaces and biomolecules is a key issue from an environmental perspective. For example, several participants of the workshop (N. Quirke and J. Subbotina) are taking part to the SmartNanotox project (<http://www.smartnanotox.eu/>) whose goal is to determine the toxicity of nanomaterials. Modeling tools are also important for determining how nanoparticles can degrade in the environment. It is also essential for modelling biominerals, such as hydroxyapatite, which forms teeth and bones, and their interaction with biomolecules in living organisms. Finally, biotechnological developments (such as biosensors, or bioanalytical devices) will often involve biomolecules coupled to solid surfaces. For example, M. Penna presented his work on antifouling coating, which are

optimised to avoid biofilm deposition that would degrade a solid surface. E. Lojou talked about redox enzymes grafted on electrodes and how they can be used to develop biofuel cells.

## 6 Participant list

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### Organizers

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**Baaden, Marc**

Institut de Biologie Physico-Chimique (IBPC), France

**Barbault, Florent**

University Paris Diderot, France

**Sacquin-Mora, Sophie**

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**Baneyx, François** - Chemical Engineering, University of Washington, USA, United States

**Bonomi, Massimiliano** - Institut Pasteur - CNRS, France

**Brancolini, Giorgia** - CNR NANO, Italy

**Chen, Zhan** - University of Michigan, USA, United States

**Ciaccafava, Alexandre** - Institut de Chimie et Biologie des Membranes & Nano-objets, UMR 5248, France

**Corni, Stefano** - CNR-NANO National Research Center, Italy

**Haynes, Christy** - University of Minnesota Department of Chemistry, College of Science and Engineering, United States

**Heinz, Hendrik** - University of Colorado Boulder, United States

**Johnston, Karen** - University of Strathclyde, United Kingdom

**Latour, Robert** - Clemson University, SC, United States

**Lojou, Elisabeth** - Bioénergétique et Ingénierie des Protéines UMR 7281, Marseille, France

**Paul, Mulheran** - Chemical and Process Engineering, University of Strathclyde, UK, United Kingdom

**Penna, Matthew** - RMIT University, Australia

**Quirke, Nick** - Imperial College London, United Kingdom

**Raffaini, Giuseppina** - Politecnico di Milano, Italy

**Sarupria, Sapna** - Clemson University, United States

**Walsh, Tiffany** - Deakin University, Australia

# Green's function methods: the next generation 4

Location: CECAM-HQ-EPFL, Lausanne, Switzerland

Webpage: <https://www.cecarn.org/workshop-details/137>

Dates: May 14, 2019 - May 17, 2019

## 1 State of the art

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Green's functions have always played a prominent role in many-body physics. In particular the one-body Green's function (GF) delivers a wealth of information about a physical system, such as ground-state energy, excitation energies, densities and other measurable quantities. Therefore, the development of approximate methods to calculate the one-body GF has been an active research topic in many-body physics since the 60's, and many routes have been explored in order to find increasingly accurate GFs. A very popular class of methods is based on the iterative solution of an integral equation for the GF containing an effective potential, the so-called self-energy, which needs to be approximated. The well-known GW approximation belongs to this class; this approximation is the method of choice for calculating band structures, but it also shows several shortcomings, such as the wrong description of satellites in photo-emission spectra, in particular in so-called strongly-correlated materials. Therefore, more refined levels of approximations are needed to keep the pace with the advances made in experiment. Recently much progress has been made in this direction both by going beyond standard methods and also exploring completely novel routes to calculate GF. A new wave of original ideas, understanding, and solutions, has pervaded the field and therefore we think it was timely to gather these new concepts in a workshop.

## 2 Major outcomes

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It is clear that state-of-the-art approximations are not sufficient anymore to describe new phenomena and emerging new physics that the advances in the experimental techniques allow us to explore. The so-called strongly correlated materials, for example, exhibit remarkable electronic and magnetic properties, such as metal-insulator transitions, half-metallicity, or unconventional superconductivity, which make them among the most attractive and versatile materials. Describing these systems requires to go beyond today's state-of-the-art approaches and represents nowadays one of the greatest challenges for condensed-matter theory. New strategies start to emerge which revisit fundamental equations and reformulate them in such a way to make clearer the physics they describe. New physical constraints are searched in order to obtain physical approximations. It is also clear that collaboration with other fields is essential, since similar problems are common to various domains and solutions might already exist. According to this, the workshop has shown four big axes of development, promising to tackle the new challenges in material science:

1. The accuracy of GW: the GW approach dates back till the 1960's but only recently a benchmark set for GW calculations has become available. Therefore, many discussions during the workshop pertained to approximations, accuracy and multiple solutions in GW calculations.
2. Beyond GW for the description of (strongly) correlated materials: self-consistency, vertex corrections, solution of the equation of motion of  $G$ , are all branches of the many-

body theory heavily explored today, especially for the description of effects that go beyond the electronic band structure, such as satellites, Hubbard bands, etc. These points were much discussed.

3. Combination of multiple approaches: the workshop showed an ever increasing combination of theoretical approaches for a variety of questions: one-particle spectroscopy, total energies, optical properties, fundamental behavior. Several topics concentrated on the combination of the following Green's function approaches: many-body perturbation theory, quantum chemistry approaches dynamical mean field theory and density-matrix functional theory.
4. Non-equilibrium theories. the workshop showed the increasing interest in developing non-equilibrium theories, for two main reasons: i) the increasing interest in thermodynamic properties, and the (dynamic) process of phase transition (like the Mott transition), often in conjunction with thermal effects; ii) the description of time-resolved spectroscopies, more and more used in the new generation synchrotron radiation facilities to investigate properties of matter, that crucially require theoretical description and analysis.

## 3 Community needs

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Many of the concepts here reported will benefit from an increase support of fundamental research. This is today particularly important in a moment in which public research is funded via contracts and projects. In many countries, fundamental research suffers with respect to applied or industrial research. Very often national calls give high priority to joint research+industry applications. To this we can add the perverse ramification of calls (European, national, regional, local) that far from multiply the offer, rather it creates a frustrating scattered and highly inefficient funding procedure. For what concerns specifically software and hardware, we have both good and bad news. Many groups have already partnerships with local software engineers, few others decided to ask support to the PRACE facilities. This is a crucial step: it is indeed true that the computer power available today (both in national and European calls) is good news to tackle bigger and more complicate systems. However, without a proper writing (and often, re-writing) of our computer codes (and this is the bad news), to take full advantage of the modern paradigms (blue-gene low memory nodes, GPUs accelerators, mics, and all hybrid architectures), there will not be any possibility to tackle such complex materials and behaviours. The scalability required to take full advantage of this new kind of resources is in fact such (1-1000000 cores) that only a professional approach can succeed. This professional procedure comes not only in term of parallelization of the codes, but also in their everyday maintenance and in all aspects of collaborative development (revision control, tickets, pull requests, forums, etc.), as previously mentioned. Finally, in order to make these new codes and capabilities widely available, training strategies have to be devised, at both hard (summer dedicated schools) and soft (e-learning platforms, MOOCS) levels.

## 4 Funding

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As mentioned before, it is crucial for the development of the fundamental research involved in this workshop, that funding agencies continue or even increase their funding of fundamental research. The workshop brought together many established experts in the field of Green's function methods as well as young promising talented researchers. At today, several channels have been analyzed, for the funding of fundamental research: - ERC grants at the European level (starting, consolidator and advanced, together with synergy) are the most important call in which fundamental theory can be funded. - Marie-Curie calls (Individual fellowship, CoFUND, and ITN) are still active and available also for fundamental research (even if some calls, useful in the past, like the career integration, are now closed) - National funding agencies are uneven (good for Germany, bad for Italy, worsening in France, etc) - local/regional calls are sometimes good sources of funding, but only for punctual needs.

## 5 Will these developments bring societal benefits?

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Progress in Green's function methods is of great importance to fundamental research. However, it already shown the crucial usefulness also for European Industry. Many-body Green's functions contain a wealth of information and this information can be used to develop novel materials for various innovating technological applications. For example, the single-particle Green's function contains information on the electron addition and removal energies, which allows for an accurate description of band gaps, band alignments etc.. This is very important information in the development of photo-voltaic devices. The single-particle Green's function also contains information on the current density, which can be used to describe accurately all kinds of phenomena related to electron transport. From the two-particle Green's function we can obtain information about the interaction between two particles such as the interaction of an excited electron with the hole it leaves behind. This is crucial information for the development of new photo-voltaic and photo-catalytic devices, for which its success largely depends on the ability of the material to split the electron and the hole after their formation. However, this applies to the field globally. Concerning the more restricted focus of the workshop in particular, it is about fundamental research preparing the long term future on a scale of, say, 10 years which unfortunately is not always of direct interest for a private company.

## 6 Participant list

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### Organizers

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**Berger, Arjan**

University Paul Sabatier, France

**Romaniello, Pina**

Université de Toulouse, France

**Sottile, Francesco**

Ecole Polytechnique, France

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**Alqahtani, Jehan** - Lancaster, United Kingdom

**Attaccalite, Claudio** - Institut Neel CNRS/UJF, Grenoble, France

**Blase, Xavier** - CNRS, France

**Bloechl, Peter E.** - Clausthal University of Technology, Germany

**Bruneval, Fabien Bruneval** - CEA, France

**Chen, Peng** - Italian Institute of Technology, Italy

**Cudazzo, Pierluigi** - Université Du Luxembourg, Luxembourg

**Dine, Khaled** - Technology Faculty - University of Saida, Algeria

**Dvorak, Marc** - Aalto University, Finland

**Georgiev, Vihar** - University of Glasgow, United Kingdom

**Gholizadehaghoyeh, Reza** - TSINGHUA UNIVERSITY, China

**Giesbertz, Klaas** - Vrije Universiteit Amsterdam, Netherlands

**Gonze, Xavier** - UCLouvain, Belgium

**Hallberg, Karen** - Instituto Bariloche, Argentina

**Held, Karsten** - Vienna University of Technology, Austria

**Loos, Pierre-Francois** - Laboratoire de Chimie et Physique Théoriques, CNRS., France

**Mansouri, Masoud** - University of the Basque Country (UPV/EHU), Spain

**Marmodoro, Alberto** - Ludwig Maximilians Universität, Germany

**Olevano, Valerio** - CNRS Institut Neel, Grenoble, France

**Onida, Giovanni** - Università degli Studi di Milano, Italy

**Reining, Lucia** - CNRS-Ecole Polytechnique, France

**Rinke, Patrick** - Aalto University, Finland

**Roedl, Claudia** - IFTO, FSU Jena, Germany

**Sahoo, Mihir Ranjan** - Harish-Chandra Research Institute, India

**Sangalli, Davide** - Consiglio Nazionale Delle Ricerche, Italy

**Sharma, Rajeshkumar** - Marwadi University, Rajkot, British Indian Ocean Territory

**Stefanucci, Gianluca** - University of Rome, Tor Vergata, Italy

**Taghizade, Narges** - Iran University of Science and Technology, Iran

**Tal, Alexey** - EPFL, Switzerland

**Tirimbo', Gianluca** - Eindhoven University of Technology, Netherlands

**Tsirkin, Stepan** - University of Zurich, Switzerland

**Vacondio, Simone** - CNR-NANO S3, Italy

**Van Setten, Michiel** - Université Catholique de Louvain-la-Neuve, Belgium

**Vanzini, Marco** - EPFL, Switzerland

**Vorwerk, Christian** - Humboldt University Berlin, Germany

**Wetherell, Jack** - University of New York, United States

# Fundamentals of density functional theory for $T > 0$ : quantum meets classical

Location: CECAM-HQ-EPFL, Lausanne, Switzerland

Webpage: <https://www.cecam.org/workshop-details/113>

Dates: May 20, 2019 - May 23, 2019

## 1 State of the art

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The original formulation of density functional theory (DFT) by Hohenberg and Kohn in 1964 was for quantum systems at zero temperature. They proved that the many-electron ground state is uniquely determined by the electron number density. In the next year, quantum DFT ("qDFT" we called it for the Workshop) was extended to non-zero temperature ( $T > 0$ ) equilibrium systems by Mermin. Elucidation of mathematical subtleties and of relationships with other ground-state many-electron formulations followed in the next 35 years and continues for  $T > 0$  today. Concurrently practical applications to electrons in periodic systems (solids) began and grew rapidly. In about 1993 explosive growth in molecular applications began as well. Most applications use the Kohn-Sham mapping of the DFT variational problem to the solution of an effective single-particle eigenvalue problem. Even today, almost all Kohn-Sham calculations are for the quantum ground state ( $T = 0\text{K}$ ) because the electron Fermi temperature is high relative to system temperatures for most solids or molecular environments. Major theoretical effort continues to be devoted to finding better exchange-correlation functionals, the only approximation in the Kohn-Sham equations. From the mid-1970s, and mostly quite independent from qDFT, the development of classical DFT ("cDFT" for the Workshop) for atomic, molecular and soft matter (e.g. colloidal and polymeric) systems also has flourished. In cDFT the basic variable is the one-body particle density. The physical systems considered correspond to temperatures for which the thermal de Broglie wavelength is much less than the mean interparticle separation. A major cDFT challenge is to develop accurate correlation free energy approximations; there is no exchange-a purely fermionic concept. This task is complicated by the fact that, distinct from typical qDFT, cDFT applications can involve multiple species, thus many different interaction potentials. Integrating out degrees of freedom to obtain effective interactions is common and this is not usually encountered in qDFT. Although the DFT literature is now exceptionally large, the overlap of the two variants, cDFT and qDFT, is almost nil. However, recent interest in previously under-explored state conditions (e.g., warm dense matter) has led the qDFT community to study electrons at much higher temperature. Unfavorable computational cost scaling of ab initio molecular dynamics for such regimes has driven renewed interest in qDFT that uses only the Kohn-Sham decomposition but not the explicit Kohn-Sham representation. This is called orbital-free DFT (OFDFT). The challenges of developing approximate functionals for OFDFT and the attendant computational issues resemble those encountered in the cDFT community. In parallel, cDFT has met new challenges for high temperature plasmas (e.g., high energy density systems) which necessarily confront the interface of classical and quantum effects. This convergence of physical challenges meant that the time was ripe to foster a serious exchange between representatives of these two communities. Among other things, this opportunity led to the deliberate structuring of the program by the organizers so that half the time allotted to each speaker was reserved for discussion.



## 2 Major outcomes

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- To provide context, overviews of cDFT (Evans) and of qDFT at both  $T=0K$  and  $T>0$  (Trickey) were presented at the outset.
- Power functional theory (Schmidt) for dynamical properties of classical systems was described.
- Relationships between cDFT and qDFT were discussed (Burke) with emphasis on exact results for the Hubbard dimer.
- Mappings between quantum and classical descriptions and exact limits were presented by Goddard.
- The key role of hard spheres as a reference system in cDFT was discussed by Roth.
- Nagy presented an orbital-free approach to  $T>0$  qDFT for systems of spherical symmetry.
- Brader developed further the power functional theory, introducing a non-equilibrium Ornstein-Zernike equation.
- Dufty discussed the relation between DFT and thermodynamics for both quantum and classical systems.
- Laird described the use of simulation to constrain and validate approximations in cDFT.
- Savin discussed the possible lack of fidelity of density functional approximations (DFAs) to the rigorous content of qDFT and showed weaknesses of the elementary statistical methods by which DFAs are tested in the qDFT literature.
- Missed opportunities in qDFT development were discussed by Dharma-wardana in terms of a “neutral pseudo-atom” picture offering new insight into electron-ion systems.
- Oettel discussed the use of machine learning to model density functionals in both quantum and classical systems. Applications on both sides were presented:
- Borgis explained how cDFT is used to study the hydration/solvation of large organic molecules in solution.
- Murillo discussed data science and  $T>0$  qDFT as routes to determining effective potentials for modelling of inertial confinement fusion and summarized results of a monumental simulation.
- Lutsko reviewed classical Fundamental Measure Theory showing density non-locality always occurs as a convolution, a significant computational advantage. This concept might be useful in quantum functional development. He presented recent work on the difficulties of applying cDFT to solid clusters.
- Egerov discussed application of cDFT to phase behavior of semi-flexible polymers in confinement.
- Sjostrom presented work aimed towards a hybrid qDFT in which core states are described with OFDFT and valence states with explicit Kohn-Sham orbitals.
- The workshop concluded with Hummel describing recent developments in  $T>0$  coupled-cluster theory (a potentially valuable benchmark for qDFT).

Open issues that were identified:

- The need for different qDFT developments to be tested on the same systems and with better statistics in order to allow direct comparisons and thereby ascertain the quality of approximations.
- The need to develop cDFT functionals that yield stable crystals.
- The use of non-local functionals (of the type that are standard in cDFT) in qDFT. Most non-local functionals in qDFT are constructed from Kohn-Sham orbitals, hence are only implicit density functionals. Is there a role for weighted density approximations, commonly employed in cDFT? (Some weighted density approaches were developed for qDFT by Gunnarsson et.al in the late 1970's but are little used.)

- The need to delineate where qDFT is necessary and where classical approximations suffice (including the validity of use of effective quantum potentials).
- The need for further development of more sophisticated quantum/classical mappings and for deeper understanding of the fundamental connections.
- The need to explore further machine learning techniques. Current work is in a very early stage for both cIDFT and qDFT and the results are a mixture of enticing prospects and under-whelming achievements.
- The exciting opportunity for developing systematic hybrid quantum/classical DFT. Here the goal would be to develop accurate functionals of both the ion and electron densities for warm-dense matter.
- The extent to which two-body correlation functions can be recovered accurately from one-body quantities such as the local particle density. cIDFT and qDFT approach this problem differently but we identified common ground that should lead to progress.

Other outcomes:

- The ‘proper’ quantum Ornstein-Zernike (QOZ) equation for the electronic case was discussed by Evans and by Dufty. The consensus that emerged from the Workshop is that we now know what the proper equation is and how QOZ is related to the widely used cIDFT treatment. Importantly, we know what is wrong with several attempts to introduce the quantum version. This is a valuable advance.
- The identification and interpretation of distinct vocabularies for essentially the same objects, e.g. “intrinsic Helmholtz functional” is the cIDFT counterpart of the qDFT “universal HK functional”. One cannot over-emphasize the need to identify a common vocabulary. Translating between the two communities is not easy but we did succeed!
- Insight as to why some formal objects are more important in one community than in the other. Example: why is the non-interacting entropy important for qDFT approximate OFDFT? cIDFT has no reason to deal separately with the entropy. Rather it focuses on the Helmholtz free energy from the outset and for the non-interacting classical system we know the corresponding functional exactly. In qDFT the leading term for the gradient expansion for weakly inhomogeneous systems shows that the non-interacting entropy has different temperature dependence from the non-interacting kinetic free energy.

## 3 Community needs

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- In cIDFT, unlike in qDFT, there are very few standard, distributable (preferably open-source) codes for doing state-of-the-art calculations.
- As the scale of simulations increases, reproducibility is beginning to emerge as an issue. The study reported by Murillo is an example; for the foreseeable future no academic could afford to repeat it or test it.
- A workshop on dynamics, transport, and hydrodynamics as related to time-dependent DFT could be useful. Most of the discussion of dynamics in this workshop was on the cIDFT side where substantial progress has been made following the development of power-functional and dynamical DFT approaches. There was rather little quantum TDDFT. Bringing together the classical and quantum dynamics DFT communities would be useful.
- Running an update of this Workshop in about 3 years could be quite useful. Sooner is probably not needed. (Aside: A problem we encountered was that the short interval between approval of our proposal and the actual meeting date meant that several people who had expressed serious interest in participating had already made conflicting calendar commitments. Advance notice seems to be tending toward one year.)

- Some informal discussion at the Workshop concerned organizing thematic or focus sessions on the cIDFT-qDFT interface at future national or international meetings. There was support for this.
- Side remark – the CECAM staff were extremely helpful in all aspects of the organization and the excellent Lausanne facilities were vital to the success of the Workshop.

## 4 Funding

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- Joint research proposals involving both European and US workers are difficult because of quite distinct differences in funding policies and practices. Thus, there was no significant discussion of such projects.
- However, we have urged all participants to call the attention of funding authorities to the Workshop topic by sending the Workshop program booklet to program managers and funding administrators.

## 5 Will these developments bring societal benefits?

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- The widespread use of qDFT codes in materials and molecular simulations for pharmaceuticals, catalysis, inertial confinement fusion processes, nuclear damage of reactor vessels, and many other applications is a major indicator of the societal impact of 55 years of qDFT research and method development. cIDFT competes with and informs (classical) MD and MC simulations which underpin a huge fraction of materials and soft matter science.
- If those impacts are any guide at all, the potential societal impact of cIDFT-qDFT hybrid methods, better effective potentials, open-source cIDFT codes or modules, more accurate and more broadly applicable functionals, undergirding formal progress, etc. also is high.

# 6 Participant list

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## Organizers

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**Dufty, James**

University of Florida, United States

**Evans, Robert**

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**Lutsko, James**

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**Trickey, Samuel**

University of Florida, United States

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**Borgis, Daniel** - CNRS, France

**Brader, Joseph** - University of Fribourg, Switzerland

**Burke, Kieron** - UC Irvine, United States

**Dharma-Wardana, Chandre** - National Research Council of Canada, Canada

**Egorov, Sergei** - UVa, United States

**Goddard, Ben** - University of Edinburgh, United Kingdom

**Hummel, Felix** - Technische Universität Wien, Institute for Theoretical Physics, Austria

**Laird, Brian** - University of Kansas, United States

**Murillo, Michael** - Michigan State University, United States

**Nagy, Agnes** - University of Debrecen, Hungary

**Oettel, Martin** - University of Tübingen, Germany

**Roth, Roland** - University of Tuebingen, Germany

**Savin, Andreas** - CNRS and Sorbonne University, France

**Schmidt, Matthias** - Universität Bayreuth, Germany

**Sjostrom, Travis** - Los Alamos National Laboratory, United States

# What about U in nanoscale systems?

Location: Zaragoza Scientific Center for Advanced Modeling (ZCAM), CECAM-ES, Zaragoza, Spain

Webpage: <https://www.cecarn.org/workshop-details/156>

Dates: May 21, 2019 - May 24, 2019

## 1 State of the art

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In the last decade an increasing effort has pushed the boundaries of the methodologies to treat electronic correlations in an ab-initio framework beyond Density Functional Theory (DFT). It would be a stretch to claim that the problem is solved, but we have now a broad spectrum of well-established methodologies (DFT+U, DFT+DMFT, DFT+Gutzwiller; GW; GW+(E)DMFT, RDMFT, etc.) and we know the pros and cons of each. Previous workshops of the “What about U?” series have contributed significantly to investigate the common theoretical framework and the differences between them, to clarify specific strengths, weaknesses, and ranges of applicability, and to stimulate cross fertilization between different approaches and approximations in order to achieve the maximum accuracy with the minimum computational effort. However, most of these methods were designed for homogeneous periodic systems. On the other hand, Materials Science is providing us with a variety of “nanoscale” systems, in which new challenges for the theoretical description, but also novel opportunities to reveal new physical phenomena, arise due to the spatial inhomogeneity. This workshop gathered scientists who started to approach this new frontier of treating strong correlations in nanoscale systems in a first-principles framework, and to showcase different phenomena arising from strong correlations in a variety of nanoscale systems. Indeed, due to the lower coordination of atoms in nanostructured materials, and the concomitant stronger localization of electrons as well as the reduced screening, electronic correlations in these systems are often enhanced compared to their bulk counterparts. Consequently, experiments with nanoscale devices have uncovered a wealth of phenomena associated with strong electronic correlations, such as Kondo effect, Coulomb blockade, non Fermi liquid behavior, and quantum phase transitions. On the other hand, strong electronic correlations are notoriously difficult to account for in ab initio electronic structure calculations and additional methodological efforts urgently need to be undertaken for a more accurate and predictive description of the above-mentioned correlation-related phenomena occurring at the nanoscale. This, in fact, was one of the main objectives of this workshop, extending the tradition of the “What about U?” series. In line with its broader scope the workshop also addressed correlation effects and their computational treatment in a large variety of nanoscale systems that are currently at the focus of intense experimental and theoretical research: (I) Supported nanoscale systems, including clusters/molecules on normal and superconducting substrates; (II) Molecular devices; (III) Layered structures, surfaces and two-dimensional materials; (IV) Strongly correlated materials with defects.

## 2 Major outcomes

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The workshop followed very closely the CECAM spirit by giving ample discussion time (talks were 30 min, followed by 15 min discussion; in addition to the round table discussion). A decisive advantage which made the workshop truly interesting was the diverse background of the speakers and participants: it was the occasion to confront density functional-based theories to many-body approaches, the quest for parameter-free descriptions to phenomenological models, quantum chemistry to solid state approaches, to argue about general strategies, identify differences in goals (spectroscopy vs. total energies) and jargon, to translate between the different languages of different communities and last but not least, to confront theoretical concepts and results to experimental findings. In the following we give a summary of the presented talks and discussions. The first talk of the workshop was given by George Sawatzky, Vancouver. Under the title "I am interested in U (don't take it personally!)", he presented a large panorama of various aspects related to screening of Coulomb interactions in solids. The origin of screening in bulk and nanosystems was scrutinized, and directly observable consequences of strong local Coulomb interactions such as multiplets in spectroscopies for f-electron materials were reviewed. Massimo Capone followed up with a pedagogical introduction to Dynamical Mean Field Theory, covering basic concepts, advantages and limitations, as well as an overview of the wide spectrum of branching applications, among others to nanoscale systems. Andrea Droghetti (UPV/EHU San Sebastian) presented his work on electronic transport through correlated molecular junctions from first principles following two different routes: (i) staying in a density-functional framework and (ii) by combination of DFT calculations with impurity solvers for treating a small strongly correlated subspace in the molecule. Interestingly, he showed that the addition of a derivative discontinuity to the LDA xc potential in realistic calculations ab initio calculations of a molecular junction allows to describe the Kondo plateau in zero-bias conductance as a function of gate voltage. Ivan Rungger's (NPL, Teddington) presentation of dynamical mean field theory calculations for piezoresistive materials for thin films for novel transistor applications demonstrated impressively how closely the field can nowadays connect to questions of practical relevance in materials science. The materials under scrutiny, SmSe and SmS, whose multiplet structures evidence the true need for a manybody description beyond DFT or DFT+methods, was discussed in comparison to the celebrated SmB<sub>6</sub>. Katharina Franke (FU Berlin) introduced her audience to the magic world of Scanning Tunneling Spectroscopy (STS) of magnetic atoms and molecules on superconducting substrates. So-called Shiba states which emerge in these systems as a direct consequence of the interaction between the magnetic moment and the superconducting states, can be measured by STS and allow to characterize magnetic states and the interaction strength with atomic-scale resolution. During the last talk of the first day, Samir Lounis (FZ Jülich) demonstrated an original combination of time-dependent density functional and many-body perturbation theory and its application to magnetic atoms on surfaces. Among other issues, he pointed out the need of an improved functional that he constructs based on sum rules and exact limits. His first principles description of zero-point fluctuations of magnetic moments in these systems provoked a heated discussion, since the persistence of magnetic moments at zero temperature was suspected to be an artifact of the method that does not describe Kondo screening. Nevertheless, it was speculated that this zero-temperature calculation might find a regime of application just slightly above the Kondo temperature, if zero point fluctuations survive up to that energy scale. The second day was opened by Robert van Leeuwen (U Jyväskylä) who introduced his work on higher order expansions of the many-body perturbation theory series beyond the GW approximation, and the pitfalls that open already at second order in  $W$ : unless special criteria for selecting diagrams (that sum up to completed squares in a way defined by Robert) are selected, such straightforward looking extensions lead to negative spectral weights. The audience was also intrigued by his finding of a careful selection of higher order diagrams finally leading to a result that comes close to a much simpler

second-Born like choice of terms, if undressed Green's functions are used. Finally, Robert introduced his audience to non-equilibrium many-body theory, preparing also the terrain for the following talk by Martin Eckstein. Martin Eckstein (FAU Erlangen-Nürnberg) showed dynamical mean field theory results within a Keldysh non-equilibrium formalism, relating to ultrafast spectroscopy experiments. An intriguing result concerned the comparison between Hartree-Fock+DMFT and GW+DMFT, which in the case investigated showed impressive agreement at equilibrium while the cheaper Hartree-Fock based scheme was clearly falling short in the out-of-equilibrium case. Gianluca Stefanucci (Rome Tor Vergata) presented a novel density functional-based description of transport, called i-DFT: in the i-DFT formalism the Hohenberg-Kohn description is augmented by an additional variable, the current, which is conjugate to the bias voltage. Gianluca made his audience appreciate the existence of an exchange bias and the possibilities of constructing explicit functionals, at least in the case of an Anderson impurity-type problem. In the following talk, Stefan Kurth (UPV/EHU San Sebastian) showed that the i-DFT formalism could be used to extract the true many body spectral function of a system from a DFT calculation in a special setup, where one of the electrodes becomes effectively decoupled (ideal STM limit). These two talks also triggered a most interesting philosophical discussion on the transferability of results based on a functional that is designed on the basis of a relatively specific physical system (single-orbital Anderson impurity problem). The opportunities opened by a relatively little costly DFT-based description were highlighted. In the afternoon session, Wolfgang Kuch (FU Berlin) surprised his audience by his report on the emergence of a cooperative spin-switching effect whose appearance in a system of molecules on a Au surface depends however on the coverage. While many aspects of the thermodynamics of such systems seem to be well understood, the mechanism of cooperative switching bears still several mysterious aspects, and is likely to further stimulate theoretical (and experimental) work. Andrew Mitchell (UC Dublin) talked on "What about U in molecules", presenting an introduction and advanced discussion on Kondo blockade effects in multi-channel systems. His audience was most astonished about his finding of the exchange coupling derived by straightforward Schrieffer-Wolff arguments to match a directly calculated value only at truly high values of U, extremely far in the strong-coupling limit. Andrew presented improved exchange couplings that are substantially smaller than the naive value, while still demonstrating that the form of the effective Hamiltonian is the expected one. The last item on the agenda of the day was a round table discussion chaired by George Sawatzky, where different aspects of downfolding to low-energy effective Hamiltonians were discussed. Andrea Droghetti took the occasion to present work on the choice of an optimized basis spanning a low-energy space, while Silke Biermann advertised the recently developed "constrained GW" technique. The discussion highlighted the different approaches inspired from solid state physics or quantum chemistry, confronting various ways of thinking about the problem. While solid state physicists usually prefer to keep the quasi-momentum  $k$  as a quantum number, defining low-energy spaces spanned by sets of single-particle states, the focus of quantum chemists on finite (and most often small) systems allows for local descriptions. Another point of discussion was on the suitable interactions to be used within effective descriptions were solid state physicists tend to try to incorporate some degree of screening already at the level of the effective interaction, while quantum chemists do not mind to work with bare interactions. These preferences are likely not merely cultural preferences but motivated by the huge effects of screening (reducing Coulomb interactions typically by an order of magnitude) in extended solid state systems. Thursday was opened by George Booth (King's College London) who presented his recent attempts of a DMET/DMFT-inspired scheme working on the basis of density matrices while still giving access to spectral properties. His scheme is based on self-consistency relations for a (chosen) number of moments of the density matrix. His first results demonstrate that at least for the case of a half-filled particle-hole symmetric Hubbard models, these moments indeed capture a fair amount of the physics, realizing e.g. a Mott transition in the infinite dimensional Hubbard model on the Bethe lattice in a similar way as DMFT. Angelo Valli (TU Vienna) presented his recent work on correlation effects in nanoscale systems, concentrating on two very different systems: small graphene flakes and the drosophila system of strong correlations and in particular of the Kondo effect in nanoscale electronics, Co

adatoms on the Cu surface. In the former, using the Nano-DMFT approach for a finite Hubbard model connected to electrodes of the graphene flake, he demonstrated an intriguing spin/valley filter behavior which depended on which C sites of the flake were connected to the electrodes. In the case of Co on Cu, using DFT+CTQMC impurity solver calculations, he showed that a Kondo effect is not easy to achieve in the theoretical description of this system, and depends strongly on the choice of parametrization of Coulomb integrals, which is highly surprising given that the Co@Cu system is considered the Kondo system par excellence. A controversial discussion on the reasons behind these findings followed. The afternoon session was opened by Steffen Backes (Ecole Polytechnique, Paris) who presented a study of the experimentally very important question of the nature of satellites (Hubbard or plasmonic) in strongly correlated materials using the GW+DMFT approach. On the examples of SrVO<sub>3</sub> and SrMoO<sub>3</sub> he showed that by artificially reducing the static part of the interaction, Hubbard bands are strongly reduced, thus leaving only the plasmon satellites. It thus turns out that the lower satellite in SrVO<sub>3</sub> is dominantly a Hubbard band, while the upper one is a mixture of both Hubbard and plasmon contributions. This is in contrast to SrMoO<sub>3</sub>, where both lower and upper satellites are a strong mixture of Hubbard and plasmonic effects. Michael Sentef (MPI Hamburg) presented work on dynamical materials engineering, and specifically on the effect of a time-dependent U on material properties within an DFT+U scheme. The up-shot was that in NiO a phase transition from charge-transfer insulator to Mott insulator can be induced by an ultrafast reduction of U in a pump-probe experiment. The last day of the conference was dedicated to the recent experimental realization of the 2D Hubbard model in magic-angle twisted bilayer graphene (MA-tBLG). These systems offer the possibility to experimentally measure the theoretically much sought-after phase diagram of the 2D Hubbard model. José Pizarro (U Bremen) first gave a nice introduction to twisted bilayer graphene and the theoretical prediction of the emergence of a flat band at magic angles, corresponding to an effective 2D Hubbard model. In the second part of his talk, he elaborated on the theoretically and computationally challenging task of computing the effective interaction U for the emergent Hubbard model. Dimitri Efetov (IFCO, Barcelona) showed beautiful experimental results from his group at IFCO in Barcelona, realizing very clean samples of MA-tBLG. Thanks to the improved quality of the samples, they were able to observe large variety of previously unobserved states, including new superconducting domes, orbital magnets and Chern insulating states.

## 3 Community needs

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Indeed, state of the art ab initio simulations of complex materials and nanostructures generally require HPC resources. Many computational codes have been developed for this task in the last decade that are often freely available and open source. The latter is a very important aspect as it facilitates the implementation of novel methods into existing codes. We consider that the outreach to other communities and especially to experimentalists is one of the keys to progress in the field. For this reason, one of the traditions of the "What about U?" series of workshops is to invite a few experimentalists and experts from neighboring fields in order to bring in different perspectives and to foster collaborations. CECAM provides an ideal platform for these exchanges and thus responds to a crucial community need.



## 4 Funding

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The meeting concentrated on the scientific questions in the field. Funding issues were not discussed in a plenary framework. Nevertheless, the workshop topic was at the heart of topics of the Psi-k-electronic structure community who actively tries to connect to possible European and national funding schemes via member contributions. It is clear that workshops such as the present one help structuring the community and prepare for answering to upcoming calls.

## 5 Will these developments bring societal benefits?

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The research discussed in the workshop was fundamental in nature. The understanding and reliable theoretical and computational description of electronic correlations in materials is one of the most challenging topics in the realm of solid state physics. Even though there are no direct economic benefits or solutions to societal problems such as sustainability to be expected from this research, the improvement of our theoretical understanding will lead to improved computational methods that allow to predict more reliably the electronic and related properties of novel materials. Better computational methods in turn are fundamental in the search for new materials with applications that may be commercialized or improve the sustainability of our economy.

## 6 Participant list

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### Organizers

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#### **Biermann, Silke**

Ecole Polytechnique, France

#### **Capone, Massimo**

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#### **Jacob, David**

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**Aldershof, Thom** - University of Queensland, Australia

**Backes, Steffen** - École Polytechnique, Paris, France

**Booth, George** - King's College London, United Kingdom

**Dine, Khaled** - Technology Faculty - University of Saida, Algeria

**Droghetti, Andrea** - University of the Basque Country, Spain

**Eckstein, Martin** - Universität Erlangen-Nürnberg, Germany

**Efetov, Dimitri** - IFCO, Barcelona, Spain

**Franke, Katarina** - Freie Universität Berlin, Germany

**Gull, Emanuel** - University of Michigan, United States

**Kuch, Wolfgang** - Freie Universität Berlin, Germany

**Kurth, Stefan** - Institute for Theoretical Physics, Free University Berlin, Germany

**Lounis, Samir** - Forschungszentrum Juelich, Germany

**Mahamadou, Seydou** - Laboratoire ITODYS, France

**Mitchell, Andrew K.** - University College Dublin, Ireland  
**Ranjan, Prabhat** - Manipal University Jaipur, British Indian Ocean Territory  
**Rungger, Ivan** - National Physical Laboratory, United Kingdom  
**Sawatzky, George** - University of British Columbia, Vancouver, Canada  
**Sentef, Michael** - MPSD CFEL Hamburg, Germany  
**Sobrino, Nahual** - DIPC / EHU, Spain  
**Stefanucci, Gianluca** - University of Rome, Tor Vergata, Italy  
**Valli, Angelo** - TU Wien, Austria  
**Van Leeuwen, Robert** - University of Jyväskylä, Finland  
**Wehling, Tim O.** - University of Bremen, Germany  
**Zgid, Dominika** - University of Michigan, Ann Arbor, United States

# Open databases integration for materials design

Location: CECAM-HQ-EPFL, Lausanne, Switzerland

Webpage: <https://www.cecama.org/workshop-details/154>

Dates: Jun 11, 2019 - Jun 14, 2019

## 1 State of the art

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Materials design has changed quite dramatically in the last few years. With the exponential growth of computer power and the development of robust first-principles electronic structure codes, large sets of calculations can be performed automatically. This is the flourishing area of high-throughput (HT) ab initio computation. The concept though simple is very powerful. HT calculations are used to create large databases (DBs) containing the calculated properties of existing and hypothetical materials. These DBs can then be intelligently interrogated and machine-learning models trained, searching for materials with desired properties and so removing the guesswork from materials design. In this framework, various open-domain DBs have appeared online (such as AFlow, the Materials Cloud, the Materials Project, NOMAD, ...). In some of those cases, a Representational State Transfer (REST) Application Program Interface (API) is available to interrogate the DB through scripts. But, so far, it is only possible to interrogate one DB at a time and the APIs are very different from one DB to another. The OPTIMADE workshop gathered all the key developers involved in these different efforts, both in Europe and in the US, as well as some of their users with the aim to continue to develop a common API. The advantage of such a solution is twofold. It allows the users to interrogate all the databases using the same query and it permits the database developers to increase their user base. The present workshop has built up on the achievements of two previous meetings held at the Lorentz Center in Leiden, Netherlands from 2016-10-24 to 2016-10-28, and at the CECAM in Lausanne, Switzerland from 2018-06-11 to 2018-06-15. Those meetings contributed to setting the foundations of the work that has been continued at the present CECAM meeting.

## 2 Major outcomes

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Building on the results achieved in our previous workshops and subsequent discussions on the OPTiMaDe mailing list, the present workshop continued to make the various DBs interoperational through the development of the OPTiMaDe API and actual implementations of it. After brief presentations (we had chosen to have elevator pitches of 2 minutes), most of the time was devoted to coding sessions and discussions. Several groups were formed (which evolved all along the workshop) targeting either some peculiar implementation (mainly focusing on the schema both on the server and the client sides), specific changes of the API (e.g. for sorting and paging), or further developments of the API (e.g. for including trajectories). After working by groups for a few hours, a general discussion would take place in which one representative of each group would present what had been achieved in the group and gather the opinion of the rest of the attendees. Then, depending on the result of the discussion, new groups would be formed in order either to continue to advance on the topic or to start a new topic. Many very fruitful discussions took place (most of the time they would also last during the breaks and even the conference dinner). Many improvements (this can be measured by

the number of pull requests in the GitHub repository of OPTiMaDe: <https://github.com/Materials-Consortia>) were adopted for the existing OPTiMaDe API. The new specifications will be finalized in the coming months. A precise timeline has been adopted aiming at the release of version 0.10 by the end of June 2019 and a release candidate for version 1.0 by the end of November 2019. The writing of a paper has been started on Overleaf and we hope to be able to submit it when the release candidate for version 1.0 is ready. The OPTiMaDe workshop can definitely be considered a success since very important improvements were achieved for the common API and its actual implementation. The COD database has a full working implementation, and several others have partially working implementations. This puts us on touch to have several OPTiMaDe implementations working before the end of the year. More importantly, very tight connections have been established between the different projects and new projects have been incorporated. The participants really appreciated the workshop and indicated that the location had been key to its success.

## 3 Community needs

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As already discussed in the section about the state of the art, materials design could clearly benefit from the common OPTiMaDe API since the latter would make it possible to interrogate all the databases using the same query. The OPTiMaDe API will also contribute to making all these data FAIR (Findable, Accessible, Interoperable, and Reusable). The effort that has been started is thus really important and could have really high impact. It should definitely be continued trying to involve as many important players as possible. For example, at the present CECAM workshop, we welcomed a developer from the OQMD database (last year, no one could attend the workshop physically due to visa issues) and a developer of the abcd tool developed in the group of Prof. G. Csanyi in Cambridge that relies on OPTiMaDe. It is important to maintain this community with a common target (the OPTiMaDe API) that will be beneficial for the community. Furthermore, the links that have been created among the different persons can also serve a different purpose. Indeed, there has already been discussion about the possibility to use the same community to define some standards. Indeed, since the meeting gathers an important number of players in the field, this number can be used as leverage to push towards the adoption of a standard. For all the reasons above, we believe that the OPTiMaDe effort should be continued. We hope that the CECAM will continue to support the organization of this series of workshops.

## 4 Funding

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Since last year, strong links have been established with the European Materials Modeling Council (<https://emmc.info>). In particular, they indicated a strong interest about the OPTiMaDe API that is clearly in line with their objectives. Various members of the OPTiMaDe community participate to some of the boards of the European Materials Modeling Council. This clearly opens some possible funding schemes at the European level. Indeed, the European Materials Modeling presently benefits from a Coordination and Support Action (CSA) from the EU which started in November 2016. There are clearly plans to submit a new proposal when an appropriate call is made. OPTiMaDe could clearly join when this happens.

## 5 Will these developments bring societal benefits?

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Materials design has clearly a great impact on society. Many new applications require specific materials with targeted properties. Furthermore, improving the efficiency of existing technologies (e.g. denser batteries, photovoltaics, faster computers, ...) also benefits from the design of materials with better properties. The development of the OPTiMaDe API can clearly contribute to making materials design easier. Indeed, the users will be able to interrogate many different databases with the same query. This will give them access to many more materials without the need to learn a different API for each database. Furthermore, machine learning methods can source information from multiple databases. The OPTiMaDe API will also avoid the replication of calculations by different databases allowing them to spend resources broadening the bounds of materials knowledge. The social benefit is thus clearly present, though it is indirect in the sense that the OPTiMaDe API is a tool that will clearly contribute to making it easier to design materials that provide benefits to society.

## 6 Participant list

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### Organizers

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# Co-evolutionary methods for the prediction and design of protein structure and interactions

Location: CECAM-HQ-EPFL, Lausanne, Switzerland

Webpage: <https://www.cecarn.org/workshop-details/138>

Dates: Jun 17, 2019 - Jun 20, 2019

## 1 State of the art

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Recent advances in genomic sequencing technologies have led to an astonishing increase in the size of protein sequence databases. This progress greatly renewed the interest in computational methods aimed at exploiting sequence datasets to explore structural and functional properties of proteins. Particularly, the analysis of correlated mutations has attracted an ever-increasing popularity in the last decade and it is currently one of the most promising tool in structural and computational biology. In a nutshell, co-evolutionary analysis relies on the observation that pairs of residues that display highly correlated mutations tend to be located in close proximity in the protein three-dimensional structure. Notably, Direct Coupling Analysis (DCA) has emerged as the most promising method to detect the truly interacting pairs of residues by efficiently disentangling direct from less-informative, mediated correlations. Since its inception, DCA has seen different methodological incarnations ranging from analytical mean-field strategies to more computationally demanding pseudo-likelihood approximations, up to full-fledged computational Boltzmann learning. The combination of the co-evolutionary information with molecular modeling techniques has already allowed the accurate determination of high-resolution protein structures, even for experimentally challenging targets such as membrane proteins, and it can be extended to RNA. Beyond purely structural studies, it has been recognized early on that correlated mutations encode multiple conformations thus paving the way to investigate evolutionary-conserved conformational dynamics. Furthermore, protein-protein interactions represent an exciting application of DCA where the simultaneous problem of predicting the interaction partners alongside the structure of the complexes pose a fascinating challenge. Last but not least, co-evolutionary techniques offer an unprecedented opportunity to use the inferred statistical models for bioengineering purposes, by generating sequences that are optimized for some design requirements while respecting all the pairwise statistical couplings imposed by evolution

## 2 Major outcomes

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Thanks to the format of the workshop, the quality of the talks and the interest of all the participants, we had frequent and productive discussions on the state of the art in the field and its future development. The major scientific points discussed were

1. The use of co-evolutionary information in integrative biology framework for investigating biomolecular structure and dynamics. Most notably, we discussed applications to membrane proteins, protein complexes and conformational transitions. We also discussed the potential of DCA-based approaches in modeling nucleic acid structure at multiple scale, ranging from RNA riboswitches to chromatin structure.

2. The most promising algorithmic developments for increasing the accuracy of co-evolutionary calculations with particular attention to approaches based on machine-learning. Furthermore, the opportunity to use DCA to improve the quality of multiple sequence alignment was also discussed.
3. How to use co-evolutionary information to reliably infer protein-protein interaction network moving beyond simple benchmark cases, such as bacterial two component signaling systems.
4. Other applications of DCA for protein design in synthetic biology applications and for the identification of epitopes in immunology.

## 3 Community needs

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At variance with other branches of computational physics/biology, the community interested in co-evolutionary analysis does not currently rely on HPC resources. This situation might change in the coming years thanks to the development of computationally-intensive machine learning algorithms and the growing interest in large scale analysis of protein-protein interactions. Collaborations with experimental groups are already quite common in the field. A clear request from the participants has been to iterate this workshop (possibly each second year). Indeed, keeping track of the rapid development of the techniques used to extract co-evolutionary information, and of new ways to use them, require frequent meetings. It must be stressed that, despite their undeniable successes, co-evolutionary approaches are still in their infancy: the first workable algorithms are about ten-years old, and novel developments and insights appear in the literature on an almost monthly basis. In this respect, it would be important to install a series of CECAM workshops at least for the next 4-6 years.

## 4 Funding

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Discussion about funding opportunities ranged from the possible proposal of European ITN networks to the submission of projects to bilateral funding schemes, such as the Lead Agency scheme between Switzerland (SNSF) and several European countries such as France, Germany, Luxembourg, Austria and others.

## 5 Will these developments bring societal benefits?

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Although these methods are just now starting to enter a more mature phase, it is not an understatement to claim that they hold great potential for application in biomedical research. As DeepMind AlphaFold has proven recently (winning CASP 13), co-evolutionary based methods are going to be routinely used for protein folding and protein-protein interaction predictions and for protein/drug design. Furthermore, some speakers highlighted the great potential of co-evolutionary approach for protein design in synthetic biology and in the characterization of immunological response. In this respect co-evolutionary analysis are likely to benefit pharmacological research and, by consequence, society and industry.



## 6 Participant list

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### Organizers

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# Theoretical and computational inorganic photochemistry: methodological developments, applications and interplay with experiments

Location: CECAM-FR-GSO

Webpage: <https://www.cecam.org/workshop-details/132>

Dates: Jun 17, 2019 - Jun 20, 2019

## 1 State of the art

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Photochemistry is central in our day-to-day life. Not surprisingly then, it has long intrigued mankind, as the underlying light-matter interaction is at the origin of many vital processes such as photosynthesis and vision for example. Thus, understanding photochemical processes is of critical importance in order to comprehend the world around us. The relevance of photochemistry also lies in the various technological applications that have been developed over the years making use of the specific chemistry and properties initiated by the population of electronically excited states upon irradiation of molecular systems or materials. Prominent examples are sensors, data storage, photovoltaics, light-emitting diodes, and phototherapy. In the past three decades or so, computational photochemistry has gained considerable credit as a tool to investigate photochemical reaction mechanisms in organic, inorganic and even biological chromophores. This reputation has been gained thanks to the concomitant growth of computational power and theoretical developments in the field of quantum chemistry. These advances allow peering beyond the traditional interpretations of photochemistry focused on vertical excitations at the Franck–Condon geometry. The exploration of other regions of the complex multidimensional potential energy surfaces is becoming routine in relatively small molecular systems, and the synergy between accurate and global static calculations and either quantum or semiclassical nonadiabatic molecular dynamics simulations has allowed major breakthroughs in the understanding of photochemical and photophysical processes. While many computational photochemical studies have been devoted to organic photoactive molecules, theoretical investigations of the photochemistry of inorganic systems such as transition metal complexes are still relatively scarce. Among the possible reasons one can cite the difficulty i) to describe accurately electronic excited states in coordination compounds, ii) to identify the excited states that are involved in the photochemical process due to the high density of electronic states present, iii) to investigate potential energy surfaces coupled by interstate and spin-orbit couplings, iv) to determine photochemical pathways evolving on these potential energy surfaces, and v) to simulate the photodynamics of such complex systems. While all these challenges are also present to some extent in organic computational photochemistry, they are in practice much more difficult to solve in metal complexes. During this workshop, examples of different ways theory can address the various challenges one is confronted with when studying photochemical processes, with special emphasis on the photochemistry of metal complexes, were presented. Critical challenges and future perspectives have also been discussed, bringing together leading researchers in both development and application of theory and computation, and experts in different experimental approaches.

## 2 Major outcomes

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Various aspects were covered during this workshop both from methodological perspectives and applications. Regarding methodologies and computational strategies, contributions on modern electronic structure methods using quantum chemical approaches adapted to the calculations of excited states were presented. In particular, methods based on the compaction of multiconfigurational wavefunctions (non-unitary similarity transformations, density matrix renormalization group) show great promises to solve the many-body problem. Several complementary contributions were also devoted to electronic structure analyses. Methods exploiting density-based indexes, transition density matrices, natural transition orbitals and charge transfer numbers were presented. They provide simple yet powerful tools to assign the state character in particular in transition metal complexes for which the high density of states makes this assignment somewhat difficult by simply looking at the interacting wave functions. The static investigation of photochemical paths was extensively discussed during the workshop. This type of approach can provide a static mechanistic picture of the most relevant relaxation pathways, even on complex inorganic systems in which excited states are coupled by interstate and spin-orbit couplings. The location of critical structures on the potential energy surfaces including minimum energy conical intersections and crossing points are necessary to rationalize the photophysical and photochemical behaviors of such systems. The importance of spin-orbit couplings was highlighted, especially in the evaluation of intersystem crossing rates. The importance of including environment effects was also discussed. Examples were given illustrating possible strong couplings between the solvent and the solute. Analysis on how fast the solvent equilibrate with the solute is also critical. The description of solid substrates was also illustrated in the context of photocatalysis on surfaces. The importance of including dynamical effects was the subject of various contributions. In particular, illustrations of non-adiabatic excited state dynamics using either wavepacket dynamics based on vibronic coupling models, or on-the-fly trajectory surface hopping dynamics including spin-orbit couplings were given. A wide range of different applications and systems were presented during this workshop. To cite just a few, systems of biological interests such as metalloproteins were discussed. Various metal complexes of interest in photoluminescent devices, as photocatalysts, as photoactivated anticancer drugs for photodynamic therapy, and in dye-sensitized solar cells were investigated. It is noteworthy that in a number of these studies, the interplay between theory and experiment is strong. The experimentalists have largely contributed to the success of this workshop by highlighting some of the difficulties to interpret their experimental data and how theoretical calculations could help them in this task. As an example, some chromium complexes displaying red or near infrared emission from their lowest doublet excited states are very challenging to study theoretically. The characterization of these doublet states is needed to rationalize the emission properties of these complexes and to optimize their photoluminescence efficiency.

## 3 Community needs

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This workshop was a first attempt to gather theoreticians and experimentalists to discuss about inorganic photochemistry and to identify critical challenges in the field. It appeared that further methodological developments are needed in order to investigate inorganic systems involved in complex photophysical and photochemical processes. And there is definitively a need from the experimentalists in the field to develop collaborations with theoreticians in order to deepen

their understanding of the photophysics and photochemistry of the complex systems under study. The format of the workshop was also very helpful, allowing ample time for discussion and interactions between the participants. The workshop gave the opportunity to bring together main players in the field and will help developing future collaborations. This meeting was timely in sharing the challenges and also new developments for addressing them in this multidisciplinary field. Based on this experience, we believe there is a strong case for a follow up meeting in several years time, which may form the basis for a regular series, which have had transformational effects in related areas of theoretical and computational chemistry some of us have been involved in e.g. non-adiabatic molecular dynamics. A second meeting would help to define specific community challenges: this first meeting showed the value of bringing together such a group.

## 4 Funding

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At this stage, there was no formal discussion about joint research proposals but several groups had the opportunity to discuss new collaborations and collaborative studies will definitely start following this workshop.

## 5 Will these developments bring societal benefits?

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The applications of the systems studied and presented in this workshop have potential social benefits. Various metal complexes of interest in photoluminescent devices (for displays, lightings and imaging), as photocatalysts (for sustainable generation of energy, photodegradation of organic pollutants and chemical catalysis), photoactivated anticancer drugs (for photodynamic therapy), and in dye-sensitized solar cells (for solar energy conversion) were investigated. One area mentioned was the possibility to replace metals that are difficult and environmentally harmful to extract with much more benign mimics, having equivalent or better functionality. The scientific progress in the field of inorganic photochemistry will certainly contribute to the developments of these impactful applications. An additional reason for a follow-up meeting in several years time would be to build new funding applications around such challenges, bringing together researchers who might not otherwise meet.

## 6 Participant list

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### Organizers

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**González, Leticia**

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# Molecular kinetics: sampling, design and machine learning

Location: CECAM-DE-MMS

Webpage: <https://www.cecam.org/workshop-details/97>

Dates: Jun 19, 2019 - Jun 21, 2019

## 1 State of the art

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The last few years have seen breakthroughs in our ability to simulate and measure the kinetics and functional mechanism of macromolecular systems such as proteins. Using special-purpose hardware or GPU clusters, direct molecular dynamics (MD) simulations can reach milliseconds. With methods based on Markov modeling, transition path sampling or hyperdynamics, kinetics and mechanisms can be simulated that involve events on the timescales of seconds and beyond. Recently, deep machine learning (ML) methods have had an impact in terms of increasing automation and accuracy of these approaches. Arguably, the main current limitation of the field of molecular kinetics theory is that it doesn't go beyond describing a given simulation setup, i.e. it doesn't yet attempt to scale, e.g. across the space of protein sequences which would be relevant to connect to the genomic revolution and to perform in silico design of functional properties such as enzyme turnover. On the other hand, ML has had significant impact in the field of quantum mechanics (QM) and material design where predicting quantities such as formation energies across chemical space is a fundamental part of the problem setting and has resulted into very sophisticated ways of learning transferable predictors that yield high accuracy and now allow QM-accuracy MD simulations. Arguably, the main current limitation of this field is that it aims at mainly predicting microscopic quantities such as potential energies or forces. The efficient and accurate computation of thermodynamic or kinetic expectation values is likely "the next big problem" faced by this community, which would benefit from input by Statistical Mechanics and MD experts. These two communities can learn and benefit from each other in order to develop new areas such as "transferable molecular kinetics" or "chemical quantum kinetics" that may facilitate the design of efficient enzymes, long-residence drugs or materials with properties that are not only a function of the crystal structure alone.

## 2 Major outcomes

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Molecular Kinetics brought together the machine learning sub-communities within both classical molecular dynamics and quantum mechanics. The workshop further attracted participants from other fields including statistical mechanics, structural bioinformatics, chemoinformatics, mathematicians and material sciences. In total the meeting saw 97 active participants. We had 21 invited speakers (5 female) who gave long form talks (30minutes + 15 minutes discussion) and short form talks (10+5 minutes) contributed junior scientists (3 thereof 1 female). Each of the speakers introduced and presented several topics in an expert manner and the cover topics included:

- approximating molecular wave-functions
- predicting properties of molecules,
- sampling large biomolecular systems

- designing new molecules with particular properties
- systematic coarse-graining
- multi-scale problems.

The majority of the talks discussed principled machine learning approaches to all of these problems. The questions session in between talks were lively and scholar covering topics but also opening up broader discussions about outstanding problems including:

- How do we improve transferability in machine learning force fields and kinetic models?
- How can we ensure the correct thermodynamics and kinetics of coarse-grained models?
- What can we expect from predictions beyond the training domain?
- How do we quantify uncertainties?

On the first day of the workshop the discussions continue until the late evening during the workshop poster session. Participants of all career stages contributed a total of 55 posters.

## 3 Community needs

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Within the represented communities there seemed to be an increasing interest and enthusiasm about transparency in methods as well as open sourcing code, data and protocols. The discussion about reproducibility was brought up several times and good intentions were backed up by actions. Since it is rare to assemble such a diverse group of people continuing the regular organization of this event will be extremely valuable to the community to facilitate the progress in the emerging use of Machine Learning methods in the molecular sciences.

## 4 Funding N/A

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## 5 Will these developments bring societal benefits?

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The importance of a quantitative understanding of molecular kinetics is increasingly recognized to be a critical component of effective drug-design. For example, effective strategies quantitative modeling of dissociation rates of ligand binding is therefore a high-priority topic at the interface between molecular kinetics and drug design. Such strategies would open up for more targeted optimization of important pharmacological and pharmacokinetic parameters. We had representatives present from two companies from the pharmaceutical industry.

## 6 Participant list N/A

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### Organizers

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# Emerging behaviour in active matter: computational challenges

Location: University of Lincoln, Lincoln, UK

Webpage: <https://www.cecarn.org/workshop-details/112>

Dates: Jun 27, 2019 - Jun 29, 2019

## 1 State of the art

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Active Matter systems, which consist of self-propelled particles that extract energy from their environment, can collectively form out-of-equilibrium stationary states that exhibit structural and/or dynamic order unseen in the equilibrium realm. Examples of such self-propelled particles in nature include flocks of birds, schools of fishes, and bacterial colonies. Due to its importance in biological and life sciences, and its promising applications, the study of active matter is a rapidly developing field at the interface of physics, mathematics, chemistry, biology and engineering. In particular, computer simulations have provided an invaluable tool to study these complex systems. Within active matter, it is convenient to distinguish between two general classes of self-propelled particles. In the first, we find systems in which the particles interact via the background through which they move (i.e., wet systems), in the second, the particles interact through kinematic constraints (imposed, for example, on their velocities), with the explicit dynamics of the background being ignored (i.e., dry systems). Examples of the first type of active matter include most biological systems, such as molecular motors, swimming microorganisms, and crawling cells; non-biological examples include so-called active or self-phoretic colloids, which use their inherent asymmetry to transform local chemical or electromagnetic energy into directed motion. This includes active colloids such as Janus particles, which catalyze distinct chemical reactions on their two hemispheres in order to generate localized concentration gradients. Their dynamics is determined by many factors, such as the size and shape of the colloids, and the relative importance of the surface activity versus the particle mobility (i.e., the viscosity of the fluid medium). The collective behavior of such systems is strikingly different from that of their non-equilibrium counterparts, exhibiting non-equilibrium phase separations, giant number fluctuations, etc. The second type of active matter includes flocks of birds or schools of fish, where the dynamics of the medium is considered of secondary importance for determining the collective properties. Such systems are typically studied through Vicsek-like models, where particles adjust their direction of motion to align with the average direction of their neighboring particles. Considerable effort has been devoted to proposing microscopic models for the particle-particle interactions that are capable of reproducing this type of dynamics. These models could have far-reaching applications, for example, as a means of controlling the large-scale behavior of ensembles of robots.

## 2 Major outcomes

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The emerging behavior of active matter is a rapidly developing field of study with many subfields, which, unfortunately, show a tendency to grow apart as the field matures and further specializes. However, despite the differences in the propulsion mechanisms used by the various types of self-propelled particles, their collective dynamics does show marked similarities. It is thus natural to attempt to develop common frameworks. With this in mind, the workshop brought together leading experts and early career researchers from several

subfields of the active matter community. The workshop succeeded in strongly encouraging the participants to interact with each other, with the aim of addressing common challenges and developing unifying methods that will allow for cross-fertilization to take place among the many subfields. We identified several specific challenges and desired outcomes within five key subfields of active matter, each of which could be modeled as wet and/or dry systems. These subfields were identified as the most suitable for the participants to make significant achievements in the future, and thus, the ones where collaborative discussions and efforts would have the highest probability of success and impact. The chosen topics were the following: - Activity, stationary states and equilibrium: Many numerical active matter models have properties bearing a striking resemblance with the (thermodynamic) equilibrated properties of effective systems. Discussions revolving around what it takes for a particular dynamic model to count as an active system giving rise to non-equilibrium thermodynamic phases were held. Being able to simulate the stationary phases over very large length scales and very long time scales appeared key to resolving the opposing viewpoints raised during these discussions.

- Active colloids / Micro swimmers: When particles are dispersed in a fluid, they interact with each other not only through direct forces, e.g., the van der Waals and electrostatic forces, but also indirectly, through the surrounding fluid. Such an indirect interaction, mediated by the host fluid is known as a hydrodynamic interaction. Several different numerical methods for resolving these interactions were presented, such as Lattice Boltzmann, Multi-Particle Collision Dynamics, and Direct Numerical Simulations of the Navier-Stokes equation. Applications to different types of active matter, and their respective pros and cons were discussed in the workshop.
- Flocking and swarming systems: Numerous types of agent-based models for flocking and swarming systems have been proposed in the literature. During the workshop, key concepts in such models were discussed using the analogy of the generalized Kuramoto model for coupled oscillators. This is useful to identify common universality classes in the different systems. A novel overarching principle was also presented to characterize activity in flocking living organisms. Computational requirements of the method were very large even for small systems and possibilities to engage with Machine Learning techniques to improve performance were discussed.
- Dynamics of cells and tissues: One of the most promising theoretical models to study the collective dynamics of cells is the so-called phase-field model. Unfortunately, the predictive power of such approaches is limited by the fact that most models are still 2D. A very challenging attempt to develop full 3D phase-field models was presented in the workshop.

## 3 Community needs

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While computational methods have been actively used in the field of biological systems, most successful examples are found in molecular biology and are based on microscopic molecular simulations. However, when targeting typical active matter systems such as cells, tissues, microorganisms, and living beings, etc., the presence of multiple length- and time-scales make such detailed microscopic models useless. Thus, alternative modeling approaches are required. This is particularly important for wet systems, where the dynamics of the background fluid must be considered. Several efficient methods to simulate these wet systems have been proposed, including the Lattice Boltzmann Method, Multi-Particle Collision Dynamics, or Direct Numerical Simulations of the Navier-Stokes equation. Unfortunately, most existing codes have been developed and maintained independently by individual research groups. This has resulted in a great deal of duplicated efforts and has made it more difficult for the groups in our community to collaborate and share or data amongst themselves. Making use of common toolkits and libraries would greatly increase the productivity and improve the reproducibility of

the community's research. To address this issue, we have in mind a project similar to the “Collaborative Computational Project for Simulations of Condensed Phases” (CCP5) specifically tailored for the Soft Matter community. In addition, it is necessary to expand our contact and communication with experimentalists in biological and engineering fields, in order to ensure that (1) our simulation efforts are not wasted and (2) that they are concentrated on answering relevant high-impact questions. A further series of CECAM workshops on this topic should be encouraged to tackle these problems and move the Soft Matter field forward.

## 4 Funding

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We have identified the following funding programs as the most promising for our future joint research projects. International funds: - The Human Frontier Science Program (HFSP): Instituted to promote international collaboration in basic research focused on the elucidation of the sophisticated and complex mechanisms of living organisms (i.e., active matter). - ERC grants (EU) National Funds: - JSPS grants (Japan) Bilateral Collaborations (Joint Research Projects and Seminars) International Joint Research Program Invitational Fellowships for Research in Japan (already awarded to Prof. Ryoichi Yamamoto for his invitation of Prof. Matthew Turner) - NHS grants (USA) - EPSRC grants (UK) Discussions during the symposium identified several possibilities for joint proposals on different topics of active matter. The symposium was beneficial in crystallizing ideas for the proposals which were already in discussion before (e.g. UK team), as well as for generating new ideas (e.g. UK – France teams).

## 5 Will these developments bring societal benefits?

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Active colloids / Micro swimmers / Flocking and swarming systems: Active matter systems such as groups of micro swimmers, birds, or fish often form swarms or flocks, and typically show complex collective behavior. Of special interest for scientific study are microscopic realizations of active matter, e.g., micro swimmers or active Janus particles, because they are particularly well suited for experiments. Understanding the collective dynamic behavior of these systems can be useful for both science and industrial applications, for example, to explain the formation of biofilms or to design targeted drug delivery systems. Active particles and micro swimmers also have the potential to be used as building blocks to create smart materials for use in novel green technologies and medical treatments. Dynamics of cells and tissue: The structure and function of cells is carefully regulated by the signals they receive from their environment. Of particular interest is the transfer of mechanical forces and stresses, which in turn are known to trigger specific bio-chemical responses inside the cell that can significantly alter their behavior, inducing changes in shape, size, motility, reorganization of the cytoskeleton, and even cell proliferation and differentiation. This last example is probably the most striking, given the bio-medical applications it promises. Carefully engineered bio-materials should allow us to control stem cell fate decisions, i.e., whether or not they divide or differentiate, and which specific cell lineage is chosen. However, before this is possible, we need to have a fundamental understanding of the interactions between the cells and with the chosen bio-material.

## 6 Participant list

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### Organizers

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# Atomistic simulations in prebiotic chemistry – a dialog between experiment and theory

Location: Sorbonne University, Pierre et Marie Curie Campus 4 Place Jussieu, 75005 Paris France

Webpage: <https://www.cecarn.org/workshop-details/101>

Dates: Jul 01, 2019 - Jul 03, 2019

## 1 State of the art

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Parallel to the astonishing information growth on prebiotic strategies reconstructing life's origin the last decade witnessed an unprecedented step forward in the computational modelling of these processes. This enabled to elaborate an "in silico" approach to the origin of informational polymers. The great advantage of computations when complementing experiments in the origin-of-life studies is that they provide information on selected single molecules and chemical reactions, whereas prebiotic experiments always work with complex mixtures, which often make the interpretation of these studies very challenging. This is the area where computational chemistry might be instrumental for experimentalists, since theory may supplement the experimentally available information with an atomic level insight into the structural aspects, electronic structure changes, energetics, spectroscopic properties and dynamic behavior of the studied systems. Since computational chemistry is still not fully recognized as a frontline method to study life's origin, the main aim of the current workshop was to overview the most recent developments in this field and to map those research areas where cooperation between experimental and theoretical research groups could be fruitful in the future. Since the previous workshop organized three years ago in the topic, a positive development can be recognized in the field towards joining experiments and theory, which was demonstrated on the current meeting by an increasing number of presentations having a combined experimental-theoretical character. This clearly demonstrates the growing need of the community for an atomic level understanding of those fundamental chemical processes that could lead to the emergence of life on our planet and elsewhere in the universe.

## 2 Major outcomes

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The three days meeting has overviewed the hottest topics of the origin-of-life research in the broadest context, including both terrestrial and extraterrestrial aspects. In total 17 invited talks were presented by prominent scientist, which addressed the following main subjects: a. Extraterrestrial synthesis of organic feedstock molecules of prebiotic relevance. In this topic major developments include the application of high-level quantum chemical calculations at the interpretation of rotational spectra collected by radio astronomy. In addition, the last few years witnessed a significant step-forward in the understanding of chemical mechanisms leading to the synthesis of simple organic molecules in the interstellar space. A special emphasis was put on ion-molecule and radical-assisted chemical processes, whose theoretical description was addressed by several speakers. In addition, exciting experimental talks have been presented on the detection of organic molecules on cometary matter, an especially promising

research area yet to be explored using atomistic simulations. b. Synthesis of prebiotic building blocks from simple organic/inorganic precursors. Several speakers have addressed the synergy between computational modeling and synthetic organic chemistry at understanding the processes which could lead to the accumulation of the precursors of the first primordial informational polymers. In this topic, in particular, major achievements were made with regard to the elementary processes leading to the synthesis of nucleic acid precursors from formamide, where quantum chemical calculations and quantum molecular dynamics simulations played a fundamental role at unraveling the atomistic details of the underlying chemistry. c. Prebiotic polymerization chemistries. Formation of oligopeptides and oligonucleotides represents a hot-topic of the origin-of-life research, which witnessed an unprecedented step-forward in the last few years both from experimental and theoretical points of view. Recent high throughput quantum molecular dynamics simulations suggested a plausible mechanism for aqueous glycine condensation reactions under extreme conditions relevant to oceanic hydrothermal vents. Further, quantum chemical calculations helped to understand the mechanism of peptide bond formation on silica surfaces. Likewise, atomistic details of the anionic ring opening polymerization of cyclic nucleotides leading to oligonucleotide sequences were also rationalized using quantum mechanical modeling. Exciting new developments related to the assembly of  $\alpha$ -aminonitriles to peptides have been reported: this topic may serve as an unlimited reservoir of mechanistic problems suitable for future atomic-scale computational studies. d. Processes leading to functional biomolecules. The last group of presentations included talks related to the RNA-world theory. In this topic a particular emphasis was put on identifying molecules that may be considered as relicts of an ancient RNA-based life, like viroids or cyclic nucleotides. Due to the high complexity of the problems, this research area has so far not been targeted by theoretical studies, but with increasing amount of available structural information on these systems it could become a potential topic for computational modeling in the future. Especially, force field based methods and QM/MM techniques seem to be potent candidates to tackle problems of this kind.

### 3 Community needs

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The main aim of the conference was to promote the cross-talk between experimentalist and theoreticians working in the field of origin-of-life research. Since this is a highly interdisciplinary field gathering physicists, chemists, biologists and planetary scientists, development of a common language is needed to facilitate cooperation between the various groups of scientists. This is possible only by organizing regular meetings in the topic. High complexity of the chemical mixtures relevant to prebiotic chemistry studies might be a limiting factor for the field's future progress. This calls for the urgent development of computational software which, besides having an accuracy necessary to adequately describe energetics of chemical reactions, enables fast and efficient treatment of complex mixtures. Albeit the predictive power of modern quantum chemical calculations and atomistic simulations is widely recognized, a communicational gap exist between theorists and experimentalist working in the origin-of-life field. A possible remedy for this problem may come from publishing methodological papers, which may bring the most frequently used theoretical approaches closer to experimentalists. Knowing the possibilities and limitations of theoretical calculations may help experimental scientist at formulating the problem to be tackled with atomistic modeling more efficiently.

## 4 Funding

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Besides national funding agencies the Templeton and Simon's Foundations (both private organizations located in the US) provide support for origin-of-life studies at an international level. Among them the Templeton Foundation sponsors strictly experimental investigations. Further funding resources are represented by NASA and ESA: they provide financial support mainly for astrobiology- and astrophysics-oriented studies. The Human Frontiers Science Program and the Volkswagen Stiftung also supports research related to prebiotic chemistry to a limited extent.

## 5 Will these developments bring societal benefits?

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Life's origin belongs to one of the most fundamental questions mankind have ever raised. Thus, visibility of the results from this research goes beyond basic science and is of distinct importance even for the general public. Press releases through main-stream media world-wide are very often in this topic and are frequently related even to purely theoretical studies. The educational impact of the origin-of-life research is also enormous. Scientists working in this field (experimentalists and theoreticians both) are often invited to give talks, seminars for the general public of the widest range of academic level starting from elementary school pupils up to professional scientists.

## 6 Participant list

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### Organizers

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# Accelerating material discovery by smart high-throughput computations

Location: University of Liverpool

Webpage: <https://www.cecarn.org/workshop-details/104>

Dates: Jul 03, 2019 - Jul 05, 2019

## 1 State of the art

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The complexity and the multi-scale nature of the relationships between a material's structure and its properties makes the design of a material with given properties one of grand scientific challenges of our times. To date, predictive calculations of material structure and properties have successfully been applied to a range of materials including cathode materials for batteries, organic photovoltaics, redox-active frameworks, metal oxides, and porous solids. Different classes of materials pose different challenges but there are some commonalities. There is a constant improvement in the predictive power and efficiency of accurate but computationally demanding methods such as Density Functional Theory (DFT). At the same time, a number of approaches that are computationally less demanding but still make useful predictions are widely used in different forms including classical force fields and machine learning approaches that work as a substitute to the quantum mechanics formalism. Fast and accurate energy calculations are at the heart of all high-throughput calculations, whether they are related to crystal structure prediction or to the evaluation of the material properties. Here machine learning methods offer an alternative top-down approach in which the material's structure and its functional properties may be deduced from the existing knowledge (databases) or/and computed characteristics related to materials performance. The progress in computational techniques and increasing available computer power facilitate the use of high-throughput methods for materials design. This in turn contributes to better understanding of structure-property relationship and allows one to optimise material's function. By their nature, high-throughput methods generate a lot of data that can be used for data mining, statistical analysis, and machine learning algorithms to improve our understanding and identify new physical trends. The ability to calculate and verify experimentally the properties promotes an increasing synergistic collaboration between experiment and theory in materials design leading to unique mutual feedback circles that accelerate the discovery of new materials.

## 2 Major outcomes

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This interdisciplinary workshop brought together experts in materials chemistry, ab initio methods, force field methods, machine learning and mathematics and facilitated knowledge transfer between different scientific communities working on finding new functional materials. Thanks to the active participation of the attendees and lively discussions, we reviewed common challenges across different material classes and discussed the opportunities and best practices in the field of computationally driven materials design. The focus was on the development of enabling tools in computational chemistry and the increasing contribution of machine learning techniques in predicting materials properties. The questions that reoccurred in talks and were addressed in panel discussions included:

- How to construct the search space beyond the obvious enumeration of options?

- How to parameterise fast chemical models such as force fields and machine learning models using first-principles data?
- How to build a robust machine learning model from a small dataset (i.e.,  $n < 1000$ )?
- How to design and select good descriptors for extended solids?
- How to validate models where comparison to experiment is not possible?
- How to make sure we can gain fundamental understanding when using ML models for property predictions?
- How to validate and benchmark different ML models? Some promising future directions were highlighted:
  - Moving from studying pristine materials to materials with defects, from ordered (e.g., crystalline) to disordered (e.g., polymeric) materials
  - Combining modelling with experiment for high-throughput characterisation in a single workflow
  - High-throughput data analysis, particularly integrated with high-throughput experiments
  - Closed-loop automation, enabled by autonomous experimental workflow driven by active learning
  - High-throughput virtual screening moving from being brute-force to being smart, facilitated for example by Bayesian optimisation or evolutionary algorithms.

Examples of using machine learning and high throughput screening computational chemistry to predict the same physical property, such as adsorption isotherms in porous materials or optical gap in organic molecules, were presented. Advantages and disadvantages of using physical and data-driven approaches and the ways of using them to complement each other were shown. Examples of tools for building hypothetical structures, for managing calculations and for performing real-time data analysis were also presented. The questions addressing the balance in the field and discussed at the meeting included:

- What is more promising – feature engineering or feature extraction by deep learning?
- What is more beneficial – a small number of accurate calculations or a large number of cheap calculations?
- What should we focus on more – the high-throughput studies of existing materials or the searches including hypothetical structures?
- What's more important for ML in materials – new algorithms or more data?
- What will produce the next big improvement in the field – new hardware or new software?

The conclusion was often that each of the two options has its merit for a certain application. As a community we should aim to harvest the advantages of multiple options and synergistically combine them. The workshop gave the opportunity for networking and provided a platform for unique learning experience for students and young researchers.

## 3 Community needs

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The need for larger curated databases of both experimental and calculated materials properties, better descriptors and representations for extended solids and better smart approaches for working with relatively small datasets were highlighted. The importance of curation of material databases was highlighted and exemplified by the success of the CSD and ICSD for experimental structures and The Materials Project for the calculated structures. For the entries that do not have a similar level of accuracy, their uncertainty should be quantified. Consistent storage and access of results across methods of computation and modalities of measurement is required. The need for going from proof-of-concept studies to applying the new tools to a wide range of material targets was highlighted. To facilitate the rollout and wide use of new approaches, the tools that are designed to be used correctly by people other than

the developers are required. The development of such tools requires time and software engineer expertise as well as deep understanding of how the tools work. The inherent difficulty in reproducing published data, even when the ML models were published alongside results, the need for benchmarking and the long-term need for systematic validation tools were discussed. To address these issues, encourage collaborations and the culture for sharing tools, communities similar to The Materials Project are required. The networking with other communities, especially applied mathematicians, data scientists, robotics experts, through the events such as CECAM workshops is important to facilitate new interdisciplinary interactions. A closed-loop feedback between the theoretical studies and synthesis and characterisation is important to accelerate the discovery of new materials. Such combined computational and experimental workflows should be encouraged through funding specific projects.

## 4 Funding

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The possibility of joint research proposals was not discussed at round table discussions. There were however discussions between the participants of joint research activities and possible future projects during the meeting. The discussions carried out during the workshop have indicated that the development of high throughput computational chemistry and data-driven methods will increase their application in both academia and industry. Thus, the importance of collaborations with industry to apply and continuously improve these tools was highlighted. It was also apparent that scientific software and associated software/data infrastructures could potentially become bottlenecks for translating new methods into deployable tools. Sustained funding for scientific / research software engineering is therefore of paramount importance. While most of the highly ambitious materials research efforts are multi-disciplinary, combining strengths in chemistry, computer science, engineering, etc., it is important that the computational chemistry and molecular modelling communities have the opportunity to advance the methodology independent of the its immediate application. Therefore, it is very important that highly technical and specialised, fundamental research efforts are funded. We expect that new ideas and collaborative projects will emerge, allowing both young and senior members of our community to apply for funding. The EPSRC, in the UK, is a clear source of funding for the development of new initiatives in materials science. Some concerns were raised about the negative impact of the political uncertainty on the future research in the UK and its future role in the Horizon Europe.

## 5 Will these developments bring societal benefits?

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The talks and round table discussions during the workshop not only showed examples of applied materials modelling but also emphasised the societal importance of the main aim of the workshop, to accelerate materials discovery. While the advances in drug discovery due to incorporation of computational screening have demonstrated by the pharmaceutical industry, the computational screening in materials science is much less common and limited to separate examples in different material classes. In this workshop we covered battery materials, thermoelectrics, materials for photo and electrochemistry applications, porous materials for separation and catalysis and many others. The development of better functional materials will bring potential societal benefits such as sustainability and economic benefits.

## 6 Participant list

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### Organizers

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**Caspary Toroker, Maytal** - Technion, Israel

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**Gavartin, Jacob** - Schrodinger Inc., United Kingdom

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# Learning the collective variables of biomolecular processes

Location: INRIA Paris

Webpage: <https://www.cecam.org/workshop-details/92>

Dates: Jul 10, 2019 - Jul 12, 2019

## 1 State of the art

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Classical molecular dynamics simulations of proteins are capable of describing biomolecular structure and dynamics at an atomistic level. This potential combined with advancements in computer hardware and algorithms has led to an ever-growing interest in simulations of increasing size and length. The interpretation of the resulting “Big Data” describing complicated multiscale molecular motion presents new challenges, as microsecond simulations may give rise to numerous complex conformational transitions that require careful statistical analysis. To characterize the process of interest, it is common practice to choose some low-dimensional molecular observables – termed collective variables (CVs) – and consider their mean or distribution (structural analysis) as well as their time evolution (dynamical analysis). In particular, CVs are used to represent the free energy landscape of the system, which reveals the relevant regions of low energy (corresponding to metastable states) as well as the barriers (accounting for transition states) between these regions. By describing the molecule’s time evolution on this free energy surface via a Langevin equation, we can directly study the pathways of a biomolecular process. Alternatively, we may employ some clustering method to identify the metastable states of the system which define a Markov state model that describes protein dynamics in terms of memory-less jumps. In a complementary way, CVs are often used in algorithms designed to enhance the sampling of rare events in simulations. For these two complementary objectives (post-processing tool to analyze molecular dynamics trajectories, or numerical tool to enhance the sampling), the construction of CVs requires a dimensionality reduction strategy. It can be achieved using linear transformations such as principal component analysis or nonlinear maps such as multidimensional scaling and diffusion maps. Recently, a variety of machine learning approaches have been proposed for this purpose. Moreover, adaptive schemes exist that iterate between exploring a CV space and using the new data to refine the reduced description. While some of those methods are mature and well-known across many disciplines, their performance depends on the specifics of the molecular process at play, the choice of the coordinate subspace, and the available statistical sampling of the data. Conversely, other methods such as deep learning have only recently been introduced in the molecular simulation field, and still developing rapidly; we still have few real-world applications to gauge their usefulness.

## 2 Major outcomes

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The workshop consisted of 5 topical sessions (in total 21 talks) on 'Dimensionality Reduction', 'Markov Models', 'Sampling', 'Machine Learning', and 'Biomolecular Processes'. Moreover, we had a poster session and 2 discussion sessions, which received major attention. Important results include the following issues: We intentionally invited scientists with quite different backgrounds, ranging from people focusing on mathematical aspects to people concerned with biochemical applications. As a result, a major outcome was to learn the terminology, the

methods, and in particular the goals of other research fields. For example, it became clear to people focusing on method development that their standard test models may be too simple to be helpful for describing biologically relevant systems (this argues in favor of the development of standardized benchmark test cases, see next section). On the other hand, people focusing on biochemical applications learned about the limitations of standard methods (e.g., the dimensionality used in clustering), given the typically very limiting sampling of molecular dynamics data. We distinguished the use of CVs for analysis (to gain insights into mechanisms) and for enhancing the sampling (as a direction describing the important features to sample, putatively difficult to sample and/or slow). Methods belonging to the two categories were presented during talks and discussed during subsequent sessions. Moreover, we acknowledged the similarity between looking for good continuous-valued CVs and discrete-valued CVs (states). In terms of mechanisms, it is not clear how to measure quantitatively the quality of CVs. The ability to correlate results with experimental data is crucial (even though this implies having also a correct force field, and not only a good CV). How to distinguish between correlation and causality? In terms of sampling, an important distinction has been whether the sampling method is exploratory in nature (i.e. allows to discover uncharted regions of the conformational landscape), or whether it allows to recover the probability distribution (free energy), or the even more challenging kinetic properties (transition paths, rates). Moreover, a distinction should also be made between energy landscapes with mainly energetic barriers or mainly entropic barriers. The scalar-valued collective variable that optimally describes the process is known and agreed upon: it is the committor function. How to characterize, or approximate it, in practice is a matter of current debate and development. Apart from more traditional linear approaches such as principal component analysis, more recent nonlinear methods of machine learning such as autoencoders have been used to identify collective variables. While the latter are potentially very powerful, it needs to be worked out what part of the problem should still be done manually (e.g., using physical and chemical intuition) and what part can be done in a black-box way. Defining the feature space, for example, remains a nontrivial problem. A related problem that does not yet have a good solution is the representation of configurations in symmetry-invariant forms. Another much discussed issue concerned the question what property of should be optimized by a dimensionality reduction method. Variance, correlation, timescale, pairwise distances, or...? This is particularly of interest since machine learning method allow to tailor these properties via several cost functions.

## 3 Community needs

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At present the development of post-simulation models is at a transition point from gathering concepts and ideas to a systematic, consistent and well-understood methodology. Similar as in more mature research fields such as electron structure theory and force field development, this requires an agreement of the community on well-defined benchmark problems, key observables of interest and clear quality assessment criteria. These goals were clearly spelled out in both discussion sessions, and all participants agreed on their importance, and first steps along these lines are currently taken. To go beyond the classical test systems, such as alanine dipeptide, we agreed in particular that we should first aim at reproducing brute force sampling of interesting problems, whether the data was obtained by long unbiased simulations (produced on Anton, for example) or by large-scale parallel architectures such as folding at home, and taking good care of preserving simulation parameters such as force field, ensemble and other conditions. Comparing to other data (experimental data, or data by sampling at a lower level of resolution, such as with coarse-grained simulations) seemed suboptimal at this stage, as errors introduced by the force field would become indistinguishable from the ones generated by the sampling algorithm. Another important aspect is that in order to characterize

the performance of different algorithms, codes implementing the latter need to be available, well-documented and if possible offered with a set of tutorials/simple use cases. Until this is achieved, we will keep using methods not because they are the best suited, but because of their availability. Making code available is also important to improve reproducibility of results. Of particular interest would be to develop a community grand challenge, as is done for protein folding and ligand binding. Blind competition in those fields has led to development and unbiased comparison of algorithm. Such an opportunity is missing in the field of numerical methods for molecular dynamics in general, and conformational sampling and CV discovery in particular. We realize that algorithms developed and used in other scientific fields (signal processing, image analysis, etc...) are of potential interest and use in the field of biomolecular simulations. A grand challenge may help draw the interest of these stakeholders. Larger events that may favor collaborations across disciplines are desirable.

## 4 Funding

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The issue of funding was not prominent in our discussions.

## 5 Will these developments bring societal benefits?

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The definition or 'learning' of collective variables represents a major cornerstone of the analysis of 'Big Data', which is central to many fields in research, development, industry and social sciences. For example, this is reflected in the fact that many of our students trained in data analysis have excellent job opportunities. The application field of Biomolecular processes is central to both basic research in Life Sciences, and applied work in Biomedical Science, Pharmacology, and Biotechnology. Current and future development of the tools discussed in the workshop will result in more efficient research workflows to discover key mechanisms in all these disciplines. This could mean making better use of the time and effort of research scientists, or even uncovering biological processes that would go altogether undetected using less sensitive approaches.

## 6 Participant list (Partial)

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### Organizers

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**Delemotte, Lucie**

KTH, Sweden

**Hénin, Jérôme**

CNRS, France

**Lelievre, Tony**

INRIA and Ecole des Ponts ParisTech, France

**Stock, Gerhard**

University of Freiburg, Germany

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**Allison, Jane R** - U Auckland, NZ, New Zealand

**Bolhuis, Peter** - University of Amsterdam, Netherlands

**Dror, Ron** - D. E. Shaw Research, New York, United States

**Ferguson, Andrew** - University of Chicago, United States

**Huang, Xuhui** - The Hong Kong University of Science and Technology, Kowloon, Hong Kong

**Hummer, Gerhard** - Max Planck Institute of Biophysics, Germany

**Keller, Bettina** - Freie Universität Berlin, Germany

**Krivov, Sergei** - Leeds University, United Kingdom

**Rodríguez García, Alejandro** - SISSA, Italy

**Rosta, Edina** - King's College London, United Kingdom

**Strodel, Birgit** - Research Centre Jülich, Germany

**Tiway, Pratyush** - University of Maryland, United States



# Dynamics of water in complex environments, bridging the gap between molecular and mesoscopic interfaces

Location: CECAM-FR-MOSER

Webpage: <https://www.cecam.org/workshop-details/96>

Dates: Jul 15, 2019 - Jul 17, 2019

## 1 State of the art

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Water is the most ubiquitous liquid in our environment. However, water is rarely found as a neat liquid and is instead usually in contact with a broad range of solutes and interfaces. This includes biomolecules and ions in biological contexts, water-mineral interfaces in geochemistry, water-air surfaces in atmospheric chemistry, water-metal interfaces for corrosion, and confined within porous materials for technological applications. The central question addressed by the workshop was thus: What is the impact of these interfaces on the properties of water, and especially on water dynamics? In the past few years new fundamental questions regarding the effects of these solutes and interfaces on the dynamics of water have arisen and despite numerous studies, no consensus has been found. The workshop focused on three in particular: 1) What is the physical origin of the effect of an interface on water dynamics? 2) What is the magnitude and spatial range of the effect induced by an interface on water dynamics? 3) How do the different water dynamical properties at an interface impact processes such as chemical reactivity and vibrational energy transport? In addition to these questions about physical behavior, additional methodological challenges have to be met, for both theoreticians and experimentalists, that can only be accomplished via collaboration between the two. Our goal was to reinforce the connection between experiments and simulations to promote the development of improved theoretical descriptions, the examination of complex systems with multiple experimental probes, and the consideration of general models of water dynamical behavior at interfaces.

## 2 Major outcomes

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A few clear themes, carried across the talks by multiple speakers, emerged from the meeting. These include: 1) The dynamics of liquids at planar surfaces present a key opportunity for testing theoretical methods by surface-sensitive spectroscopy. An important outcome here is the importance of simulations that directly predict the spectroscopic observable. Experimentalists at the meeting [T. Tahara (RIKEN, JP); S. Roke (EPFL, SUI); H. de Aguiar (ENS, FR); H. Allen (Ohio State, USA); N. Dupertuis (EPFL, SUI)] showed a range of new/developing spectroscopic techniques and complex interfaces that will continue to challenge theoretical descriptions. At the same time, theoretical speakers [A. Morita (Tokohu, JP), J. Hynes (U. of Colorado, USA; ENS, FR), J. Eaves (U. of Colorado); M. Baer (PNNL, USA), M. Meuwly (U. Basel, SUI), R. Rey (U. Catalunya, ESP)] showed the improving power of simulation for understanding dynamics at aqueous interfaces. 2) Open and developing

questions about the role of water in biology were discussed by a number of speakers [P. Jungwirth (Acad. Sci. Czech Repub.), T. Elsaesser (Max Born Inst., GER), P. Rossky (Rice U., USA), K. Mitchell-Koch (Wichita St. U., USA), E. Duboue-Dijon (IBPC, FR)]. A particular point of discussion involved the effect of the interface on the water dynamics (and vice versa). There is still disagreement in this area about the range of structural and dynamical correlations; this discussion is progressing towards some resolution but will require additional collaboration between theorists and experimentalists. A related question is the degree to which water near biomolecules represents a separate "species." Evidence at the workshop indicates the answer is "no" but this is not yet fully accepted in the field.<sup>3</sup>) The dynamics of and around ions in water was also substantially discussed. A subtheme was the long-standing question of the nature of a proton in water as either a Zundel ( $\text{H}_5\text{O}_2^+$ ) species or Eigen (solvated  $\text{H}_3\text{O}^+$ ). This discussion [T. Elsaesser (Max Born Inst., GER), D. Laage (ENS, FR), E. Pines (Ben-Gurion, ISR), E. Nibbering (Max Born Inst., GER), M. Johnson (Yale, USA), S. Corcelli (U. Notre Dame, USA)] which spanned the gas and aqueous phases, did not reach a resolution, but new directions for answering this question were presented. This lies in the broader context of the effect of ions on water dynamics [G. Stirnemann (IBPC, FR), A. Hassanali (ICTP, ITA), W. Thompson (U. Kansas, USA)]. In addition to physically interesting results, the discussion touched on how the phenomena can be misinterpreted and new analyses are required to uncover the driving forces which are shared with the planar and biological surfaces mentioned above.

## 3 Community needs

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Regarding the computational infrastructure, many of the phenomena of water dynamics at interfaces is accessible by classical MD simulations for which codes and significant HPC resources are available. Some of the problems, e.g., the Zundel-Eigen issue are not easily resolved without ab initio MD approaches and/or nuclear quantum effects. These are computationally challenging and indeed limiting. Some new techniques are required, S. Corcelli (U. of Notre Dame, USA) presented one such, to overcome these issues. There was unanimous agreement among the participants that the progress in this field will come only through direct, continuous collaboration between theorists and experimentalists. The inclusion of both experimentalists and theorists was viewed as a major strength of the workshop. While other conferences exist on related topics (e.g. Gordon Research Conference, Time-Resolved Vibrational Spectroscopy series), there was a unanimous agreement among the participants that the format of such a CECAM workshop makes it unique. The limited number of participants and the time allowed for discussions both favor fruitful exchanges. The general opinion was thus that such a meeting would deserve to be organized again in the future (even though not necessarily as a regular series).

## 4 Funding

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The works presented by the participants were typically funded by individual grants (e.g. NSF, DOE, ERC, JSPS). The discussions held during the workshop, especially between theoreticians and experimentalists, are anticipated to lead to future collaborations, but the issue of funding was not directly addressed at this point.

## 5 Will these developments bring societal benefits?

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While the meeting focused on fundamental questions related to the dynamics of water at interfaces, the speakers clearly indicated how new knowledge in this area will ultimately lead to benefits to society. For example, J. Eaves (Univ. of Colorado, USA) discussed how water dynamics next to a hydrophobic interface can be important in designing new paradigms for water purification. A. Morita (Tokohu U., JP) presented a theory of ion transport across water-oil interfaces that is important for understanding and designing methods for separations, sensors, and phase-transfer catalysts. H. Allen (Ohio State U., USA) presented results on air/aqueous salt interfaces that are important in chemistry of the atmosphere. P. Rosicky (Rice U., USA) provided an overarching picture of the connections between water behavior at interfaces and biological function.

## 6 Participant list

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### Organizers

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**De Aguiar, Hilton**

Ecole Normale Supérieure/Paris, France

**Laage, Damien**

Ecole Normale Supérieure, PSL Univ., Sorbonne Univ., CNRS, France

**Thompson, Ward H.**

University of Kansas, Lawrence, United States

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**Allen, H.c.** - Ohio State University, United States

**Baer, Marcel** - Pacific Northwest National Laboratory, United States

**Corcelli, Steve A.** - Department of Chemistry and Biochemistry, University of Notre Dame, Notre Dame, IN 46556, United States

**Eaves, Joel** - University of Colorado, Boulder, United States

**Elsaesser, Thomas** - Max Born Institut, Berlin, Germany

**Hassanali, Ali** - ICTP, Italy

**Hynes, James** - ENS, Paris/Univ Colorado, Boulder, France

**Johnson, Mark** - Yale, United States

**Jungwirth, P.** - Academy of Sciences of the Czech Republic, Czech Republic

**Meuwly, Markus** - University of Basel, Switzerland

**Mitchell-Koch, Katie** - Wichita State University, United States

**Morita, Akihiro** - Tohoku University, Japan

**Nibbering, Erik T. J.** - Max Born Institut, Berlin, Germany

**Pines, Ehud** - Department of Chemistry, Ben-Gurion University of the Negev, Israel

**Rey, Rossend** - Univ. Politecnica Catalunya, Spain

**Roke, S.** - EPFL Switzerland, Switzerland

**Rosicky, Peter** - Rice University, United States

**Tahara, Tahei** - RIKEN, Japan

# Fabrication processes and molecular organization in organic thin films: theory and simulation meet experiments

Location: CECAM-IT-SIMUL, Politecnico di Milano, Polo Territoriale di Lecco, Via G. Previati, 23900 Lecco, Italy

Webpage: <https://www.cecam.org/workshop-details/140>

Dates: Jul 17, 2019 - Jul 20, 2019

## 1 State of the art

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The computational study and design of soft, functional organic materials (typically pi-conjugated molecules and polymers), has made huge progresses in the last twenty years. However, there are still very significant challenges for the simulation of their behaviour in thin films, which are relevant for many applications (e.g., organic transistors, sensors, photovoltaics, etc.). The aim of the workshop was to bring together computational scientists, theorists and experimentalists, to define the state of the art and push forward the boundaries in the field of organic thin films. In particular, the following key questions were identified:

- What are the criteria for validating a simulation of an organic thin film, when the film itself may be in some non-equilibrium metastable state and information about its structure incomplete?
- What level of coarse-graining and scale-bridging strategies can make the simulations of organic thin film production processes feasible and closer to experiment?
- How can simulations be made more relevant to the interpretation of experiments? How can experimental information be used to constrain the outcome of the numerical simulations?
- What level of detail in a model is required to reproduce or predict specific properties of the films (e.g., charge transport)?
- What are the best techniques for extending simulations from the study of equilibrium molecular organizations to that of nonequilibrium ones produced by the processing techniques practically used in technology?

## 2 Major outcomes

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The workshop addressed several systems and themes in the following half-day sessions: \* small-molecule thin films: structure, properties and self-assembly processes (talks by A. Sassella, R. Resel, L. Muccioli, S. Conti) \* polymers: structure and phase behaviour (talks by N. Stingelin, M. Caironi, S. V. Meille, G. Rutledge) \* devices based on liquid crystalline elastomers and biosensors (talks by D. Broer, M. Warner, L. Torsi and F. Biscarini) \* organic electronics: structure and charge transport (talks by A. Troisi, S. Manoj, A. De Nicola) \* evaporation processes and structure formation organic in thin films (talks by M. Yoneya, A. Sevink, M. Doi). An extended poster session allowed a number of young scientists (typically PhD students and postdocs) to present their results (the one by M. Dreher was awarded a "best poster prize", following the recommendations of a small panel of senior participants).

During the discussions, which took place after each talk and in the final round table session, a number of cross-themes, trends and open problems were identified:

- simulation of out-of-equilibrium systems and self-assembly under non-equilibrium conditions;
- bridging molecular and mesoscopic scales in the simulation of evaporation and spin-coating processes;
- simulation of complex mixtures and heterogeneous systems;
- use of artificial intelligence techniques;
- validation of force fields and of simulation protocols;
- criteria for coarse-graining of the interactions and of the equations of motions;
- strategies for using experimental information to drive and validate the simulations;
- sharing and standardization of data, simulation protocols, codes.

## 3 Community needs

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Compared to other molecular systems, the simulation of organic thin films is very complex and time-consuming. A state-of-the-art simulation of an organic thin film (for a transistor, for example) involves several complex steps and decisions, in addition to significant computational resources. For example: choice of a force field (non-trivial, when organic system interacts with an inorganic substrate), definition of the simulation protocol (e.g., composition, temperature, deposition or evaporation rates, etc.), extraction of interesting film properties (e.g., order parameters, optical or electronic properties), comparison with available experimental data (e.g., GISAXS for structure and charge mobility measurements). All this typically requires a collaboration between several people and groups with different expertise. In a certain sense, these simulations are by necessity a "community effort". There are instances in which the same types of systems has been simulated by different groups (e.g., photovoltaic blends of P3HT and PCBM, or polymer nanocomposites), with different and sometimes contradictory outcomes. The cross-validation and comparison of simulation results by different groups would be beneficial, but it seems also very challenging (much more, for example, than a calculation of the electronic structure of a molecule or crystalline material). It seems unwise to attempt the definition of standard simulation protocols. However, it might be beneficial share simulation data after publication, encouraging the use of existing or new repositories.

## 4 Funding

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Research funding opportunities were not explicitly addressed during the open discussion sessions, but as organizers we are aware that they were discussed informally by workshop participants. Horizon 2020 offers many opportunities in this respect. Smaller-scale international collaborations may be obtain funding through visitor exchange programs and joint applications for supercomputer resources (e.g., PRACE calls). The conference sponsor (Lampre SpA) also invited the participants to share ideas for possible joint research projects on organic functional coatings and coating processes.

## 5 Will these developments bring societal benefits?

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Devices based on thin films of organic functional molecules have a great potential for specialized application. Examples which fall completely outside the realm of traditional (e.g., silicon-based) electronics include ultra-sensitive and specific biosensors (talks by Torsi and Biscarini) as well as "edible electronics" (talk by Caironi), which may help to achieve the goals of personalized medicine. These applications require careful experimental characterization of the device structure. In turn, the experimental results obtained from them can only be interpreted with the help of molecular simulations. Molecular simulation of organic thin films has made major progress but it is still far from the point where it can be used as a standardized, "black box" for the investigation of their structure and properties. However, there is increasing awareness in the industry that these tools will become increasingly important in the future, as testified by the sponsorship of this workshop by Lampre SpA.

## 6 Participant list

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### Organizers

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#### **Milano, Giuseppe**

Dept. Of Organic Materials Science, Yamagata University, Italy

#### **Raos, Guido**

Politecnico di Milano, Italy

#### **Zannoni, Claudio**

Universita' di Bologna, Dipartimento di Chimica Industriale "Toso Montanari", Italy

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**Baggioli, Alberto** - Politecnico di Milano, Italy

**Baumeier, Bjoern** - Eindhoven University of Technology, Netherlands

**Beljonne, David** - University of Mons, Belgium

**Biscarini, Fabio Biscarini** - Università di Modena e Reggio Emilia, Italy

**Broer, Dirk** - Eindhoven University of Technology, Netherlands

**Caironi, Mario** - IIT, CNST@PoliMi, Milano, Italy

**Conti, Simone** - Harvard University, United States

**De Nicola, Antonio** - Department of Organic Materials Science Yamagata University, Japan

**Doi, Masao** - Center of Soft Matter and Its Applications, Beihang University, China

**Famulari, Antonino** - Politecnico di Milano, Italy

**Gertsen, Anders S.** - Technical University of Denmark, Denmark

**Hofer, Sebastian** - Institute of Solid State Physics, Graz University of Technology, Austria

**Meille, Stefano Valdo** - Politecnico di Milano, Italy

**Muccioli, Luca** - University of Bologna, Italy

**Pasquini, Marta** - Politecnico di Milano, Italy

**Resel, Roland** - Graz University of Technology, Austria


**Rutledge, Gregory** - Massachusetts Institute of Technology (MIT), United States

**Sassella, Adele** - Università di Milano - Bicocca, Italy

**Sevink, Geert Jan Agur** - Leiden University, Netherlands

**Sørensen, Michael Korning** - Technical University of Denmark, Denmark

**Stingelin, Natalie** - Imperial College London, UK and Georgia Tech, United States

**Torsi, Luisa** -  Università degli Studi di Bari "Aldo Moro", Italy

**Troisi, Alessandro** - University of Liverpool, United Kingdom

**Warner, Mark** - University of Cambridge, United Kingdom

**Yoneya, Makoto** - National Institute of Advanced Industrial Science and Technology (AIST),  
Japan

# High density DNA arrays: models, theories and multiscale simulations

Location: National Institute of Chemistry, Ljubljana, Slovenia

Webpage: <https://www.cecarn.org/workshop-details/106>

Dates: Jul 24, 2019 - Jul 26, 2019

## 1 State of the art

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Densely packed DNA arrays exhibit hexagonal and orthorhombic local packings, as well as a weakly first order transition between them, as affected also by the length of the DNA strands or fragments. While we have some understanding of the interactions between DNA molecules in aqueous ionic solutions, the structural details of its ordered phases and the mechanism governing the respective phase transitions between them remain less well understood. Since at high DNA densities, i.e., small interaxial spacings, one can neglect neither the atomic details of the interacting macromolecular surfaces nor the atomic details of the intervening ionic solution, the atomistic resolution is a sine qua non to properly describe and analyze the interactions between DNA molecules. In fact, in order to properly understand the details of the observed osmotic equation of state, one needs to implement multiple levels organization, spanning the range from the molecular order of DNA itself, the possible ordering of counterions, and then all the way to the induced molecular ordering of the aqueous solvent and then mesoscopic level, all coupled together by electrostatic, steric, thermal and direct hydrogen-bonding interactions. Multiscale approaches coupling atomistic, mesoscopic, and macroscopic levels of detail therefore appear as singularly suited to connect the microscopic details of this system with its macroscopic thermodynamic behavior. With the proposed workshop, we wished to address the simulation approaches to the dense DNA arrays with different local packing symmetries, the solvent structural order, and counterion types. The aim of the meeting was to discuss numerical and theoretical methods and models at different length scales applicable to such systems to analyze the osmotic equation of state, to identify the most important contributions to the DNA-DNA interactions at high DNA densities, to determine possible liquid crystalline ordering profiles and explore means for controlling the DNA packing and ordering. In an unrestrained atmosphere of an interdisciplinary workshop including physicists, chemists, and biologists these goals may be reached to mutual benefit of the participants as well as indirectly of the scientific community.

## 2 Major outcomes

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At the workshop, we have addressed various main open issues and discussed the possible new directions of research that would extend the range of phenomena that we can currently understand by different modeling techniques:

1. we scrutinized the various multiscale methods coupling different models at various length scales and their possible relevance for different types of phenomena on spatial as well as temporal scales
2. performing open grand-canonical molecular simulations that exchange energy, momentum, and matter with the external environment as these are the most relevant formulations that can approach the existing experimental approaches to the DNA



matter performed typically at fixed ionic composition and set osmotic pressure of the aqueous solvent

3. determination of phase transitions between DNA subphases beyond invoking the general Lindemann criterion, using enhanced sampling techniques in order to elucidate the details and experimentally unresolved features-characteristics of these phase transitions
4. determination of DNA to surface interactions (surface anchoring) in view of orientational and positional order of DNA, specifically in the context of viruses and the DNA packaging the proteinaceous shells
5. identification of material regimes and system parameters for use of dense DNA materials as advanced macroscopic materials, such as in bio-photonics, which are intimately connected with a solid computational grip of the phenomena that can predict and guide the experimental realizations.

We have published a workshop booklet (attached) with abstracts. There has been 19 invited speakers and in total 28 registered participants from 11 countries participating at the workshop. Local students also participated in some activities.

## 3 Community needs

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The community needs efficient multiscale software packages (codes) coupling different methods for separate length scales in biomolecular systems as well as the access to the HPC resources to treat vast amount of data coming from e.g, the cryo-em microscopy.

## 4 Funding

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The workshop was founded by a gracious donation of 12.000 EUR from the Austrian CECAM-AT node. There have been discussions during the meeting on a possible ITN network on the topic, which will lead to a common application. There have been also discussions on a possible joint CECAM activities with SISSA initiated, which has led to a get-to-know meeting between computational researchers from SISSA (Trieste) and Ljubljana in Postojna in October (<https://www.cmm.ki.si/CECAM/>). Future workshops might be also founded by the Ministry of Education, Science and Sport, Slovenia.

## 5 Will these developments bring societal benefits?

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The workshop has had the IUPAP Conference Endorsement and has been a satellite meeting of the Joint 12th EBSA 10th ICBP-IUPAP Biophysics Congress, Madrid, Spain. We have therefore subscribed to the statement on free circulation of scientists in addition to CECAM's own policies. Apart from that, it was very beneficial for the local scientific community (theoretical and experimental) because several students as well as researchers from nearby academic institutions, e.g., Jozef Stefan Institute, National Institute of Chemistry, Faculty of Mathematics and Physics were able to attend the talks and discussions at the workshop.

## 6 Participant list

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### Organizers

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**Podgornik, Rudolf**

University of Chinese Academy of Sciences, Beijing, China

**Praprotnik, Matej**

National Institute of Chemistry, Slovenia

**Ravnik, Miha**

University of Ljubljana, Slovenia

**Ziherl, Primoz**

University of Ljubljana, Slovenia

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**Abrams, Cameron F.** - Drexel University, United States

**Adzic, Natasa** - Faculty of Physics, University of Vienna, Austria

**Aksimentiev, Aleksei** - University of Illinois at Urbana-Champaign, United States

**Bellini, Tommaso** - University of Milan, Italy

**Cremer, Christoph** - Institute of Molecular Biology (IMB), Germany

**Dobnikar, Jure** - Chinese Academy of Sciences, China

**Doye, Jonathan** - University of Oxford, United Kingdom

**Drevenšek Olenik, Irena** - Faculty of Mathematics and Physics, University of Ljubljana, Slovenia

**Giacometti, Achille** - Dip. Chimica Fisica, Univ. Venezia, Italy

**Holm, Christian** - University of Stuttgart, Germany

**Kirmizialtin, Serdal** - New York University Abu Dhabi, United Arab Emirates

**Kornyshev, Alexei** - Imperial College London, United Kingdom

**Kremer, Kurt** - Max Planck Institute for Polymer Research, Germany

**Leforestier, Amélie** - Université Paris-Sud, France

**Losdorfer Bozic, Anze** - Jozef Stefan Institute, Slovenia

**Lyubartsev, Alexander** - Stockholm University, Sweden

**Mebwe Pachong, Stanard** - Max Planck Institute for Polymer Sciences, Germany

**Merzel, Franci** - National Institute of Chemistry, Slovenia

**Micheletti, Cristian** - SISSA, Italy

**Pagonabarraga, Ignacio** - CECAM HQ, Switzerland

**Rosa, Angelo** - Scuola Internazionale Superiore di Studi Avanzati (SISSA), Italy

**Srivastava, Amit** - New York University, United Arab Emirates

**Svenšek, Daniel** - Faculty of Mathematics and Physics, University of Ljubljana, Slovenia

**Zavadlav, Julija Zavadlav** - Technical University of Munich, Germany

# Modeling noncovalent interactions in supra/bio catalysis

Location: CECAM-FR-MOSER

Webpage: <https://www.cecarn.org/workshop-details/146>

Dates: Aug 26, 2019 - Aug 30, 2019

## Major Outcomes

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The workshop started on Monday, August 26th, 2019 with the intervention of Giovanni Bistonni. He describes the Local Energy Decomposition scheme which allows to retrieve and analyze weak interactions within molecular systems. To emphasize the usefulness of this newly developed methodology, he shows an application in the domain of counter-anion catalysis, and then an extension to biological systems. He pinpoints the fact that solvent-solute interactions is still difficult to describe and analyze in terms of weak interactions: "Methods are there, but too much data, thus we need automatization". The next speaker was Qian Peng, with a talk on non-covalent interactions (NCI) in the context of homogenous organometallic reactions. He showed it is possible to derive a fitting procedure from calculations and then to synthesize new ligands inducing better ee%. On Tuesday morning, Paul Popelier gives us a background of QTAIM and other various topological analyses. Then he introduces Interacting Quantum Atoms and gives as an example water decomposition. The main message was: "Chemistry is based on cancellation of strong physical effects and thus is the results of tiny modifications". Then he gives a nice example of energetic decomposition for the understanding of interactions in HIV-1 protease. He was followed by Esmail Alikhani, presenting a talk about topology and non-covalent interactions, and summarizing history of classifications: how we build the "zoo" of weak interactions. He then gives details on how to retrieve structural information from topological analysis. Emilie-Laure Zins talks about water-solute interactions and reminds us the importance of having access to high-quality spectroscopies for understanding weak interactions (importance of theory-experience interplay). In the afternoon, Hendrik Zipse introduces radical species influenced by hydrogen bonding: it destabilizes very slightly radical oxygen species but it is the opposite for radical oxygen aromatic species. Then he demonstrates how to generate radicals starting from closed-shell species with the help of hydrogen bonding. He is followed by Jan Schümann who is discussing the balance between dispersion and non-covalent interactions. He points out that there is problem with the IUPAC definition of steric interaction/repulsion. He then talks about laderranes and the building of molecular balances. He demonstrates it is possible to match experiments and theoretical calculations with these newly introduced molecular balances. He finishes his talk by showing (again!) a problem with solvent effects. On Wednesday, the day is starting with G.N. Sastry. He is summarizing first the history of non-covalent interactions, then moving to an important problem based on NCI: kinase inhibitors (p38 MAP more specifically). The talk tries to signify that there is a fundamental difference in the way covalent and non-covalent interactions manifests in the nature, and looks at the fundamental differences between these two types of interactions. He gives us also a nice review about cooperativity in the framework on NCI. He suggests to use better force fields for scoring after docking procedure, by carefully considering factors such as polarizability, quantum refinement and cooperativity phenomenon. Ana-Rita Calixto is now talking about phosphate hydrolysis, a central problem of biology, highly sensitive to electrostatic environment. She demonstrates that EVB is able to dissect enzymatic reaction to reconstitute the role of each residues. To finish the morning, Bruno Madebene is introducing calculations on nanoparticles coated by organic substrates (namely thiolate) and studies made on them by various topological analysis, thus proving that NCI can be found in original objects.

(Afternoon: free time in Paris) On Thursday morning, Frédéric Célerse summarizes SMD (steered molecular dynamics) and its use in the context of finding rare events, more specifically with polarizable force fields. He demonstrates that non-polarizable FF are not able to describe salt bridges in a correct manner (dissociation is too easy). This point is coupled to solvent reorganization which is poorly described by classical FF. Then, Etienne Derat is summarizing organometallic reactivity inside cyclodextrins. He demonstrates how these original structures are acting like enzymes by stabilizing intermediates through specific NCI, but he points out the difficulty to describe these species with tools conceived mainly for proteins. Sugars are important in biology too and needs to be taken into account while developing simulation methodologies for biological purposes! R.B. Sunoj is then discussing asymmetric induction and how weak interactions are controlling selectivity in organocatalysis. He is showing an example of prediction in organometallic catalysis, based on clear understanding of NCI. He points out the importance of publishing negative results like low ee%, and to simulate these “bad” results to explain them. In the afternoon, Dan Major is talking about enzymatic catalysis but also catalysis made in nano-capsules. He starts by reminding us that there is sometimes artifacts in experimental data. He is continuing with the problem of QM/MM energetics convergence and describes what is needed for good quality energies (mainly cutting in a reasonable manner!). Then he discusses about terpene synthesis and how enzymes control the selectivity by giving an example of TS bifurcation controlled by the enzymatic environment. Then he shows that NCI are controlling product distribution for terpene formation in nano-capsules, the effects are weak but it is possible to capture them by simulation. Sam de Visser is finishing the day by discussing the influence of the environment on the reactivity of various metallo-enzymes presenting an oxidative species. He presents us results regarding a new type of enzyme: P450 peroxygenase, which is able to perform desaturation. He then moves by explaining how halogenases are controlling product selectivity by substrate positioning and shows how simple modeling by electric field can reproduce more elaborated multiscale simulation. He finishes with calculations on prolyl hydroxylases, where this positioning is managed by aromatic residues. The last day of the workshop is starting with Majdi Hochlaf who is describing computational strategies to retrieve interactions energies with a good ratio time/quality. He then presents the results of a double-blind challenge for solvation of basic compound (yes still a problem in 2019!). He then moves to studies of gold surface – imidazole -CO<sub>2</sub> and goes step by step to more complex system with similar reactivity (namely MOF). Nohad Gresh is then talking about polarizable force field (SIBFA), the goal being of having MD with the quality of QM calculations. He describes how this FF is built and moves to zinc proteins to show applications. He points out that water can be highly polarized in zinc active sites, which is the reason why we need high quality FF. Fernanda Duarte starts the last talk by reminding us why the accuracy is important for simulating molecular processes with selected examples. Then she moves to supramolecular catalysis and points out that despite years of experimental works, very few examples exist (Two were presented in this workshop, lucky us!). She is also talking about the difficulty to parametrize metals in FF and second sphere of solvation, even in our days. We need also better models to deal with entropy in the framework of supramolecular catalysis. After this talk, the participants started a discussion to summarize workshop main points. Here they are:

- There is no catalysis if there are no non-covalent interactions.
- Solvent is a main component of a NCI system.
- Negative results are important!
- Perception of NCI is rapidly evolving (classification), but experimentalists are also now grabbing the tool.
- Computational challenges at different length scales and time scales arise largely due to the demands in accurately depicting and quantifying non-covalent interactions in supra-molecular architectures.
- Universality of NCI from materials to biocatalysis!

Thus, from a theoretical perspective, there is overlapping problems, which can be tackle by synergy between exp/theo. Modeling is the tool of choice for interpretation, but we need a lot

of methodological development. To cite a few: database of NCI, solvent coupling to catalyst, metals & sugars in FF, entropy properly taken into account at reasonable computational cost.

## 6 Participant list N/A

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### Organizers

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**Alikhani, Esmail**

Sorbonne Université, France

**Derat, Etienne**

Sorbonne Université, France

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# New methods in Monte Carlo simulations: parallel, adaptive, irreversible

Location: CECAM-HQ-EPFL, Lausanne, Switzerland

Webpage: <https://www.cecarn.org/workshop-details/151>

Dates: Sep 02, 2019 - Sep 04, 2019

## 1 State of the art

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The Monte Carlo method is well established and broadly used in a wide range of disciplines. The Metropolis-Hastings algorithm provides a nearly universal solution to the problem of importance sampling. Cluster algorithms have been devised to beat critical slowing down at continuous phase transitions, multicanonical simulations allow to study phase coexistence at first-order transitions. These and further successes notwithstanding, significant challenges remain and the field has continued to develop in a dynamic fashion. Recent exciting advances in simulation methods include improvements to the Markov chain method as the workhorse of Monte Carlo simulations, such as event-chain techniques, fast methods for treating systems with long-range interactions, simulations replacing the diffusive motion implied by detailed balance with a semi-ballistic movement achieved through lifting, and perfect sampling ensured through coupling-from-the-past. Other approaches such as population annealing dispose with the concept of Markov chains altogether and achieve improved equilibration together with near perfect parallel scalability through the use of sequential Monte Carlo. This workshop provided an overview of the state-of-the-art in Monte Carlo simulations in statistical physics, identified the open challenges in the field, and worked towards their solution through intense discussions and exchange of ideas between researchers working on different strands of methods.

## 2 Major outcomes

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The workshop included a moderated discussion kick-started by keynote presentations of several leading researchers in the field that was found to be particularly useful for identifying the main development lines and challenges for the future. From the various formal and informal discussions there emerged a consensus that it is useful to view the typical Monte Carlo method as a combination of a "driver" method propagating the elementary degrees of freedom with a "generalized ensemble" algorithm that allows the driver technique to more fully or quickly explore the phase space of the system. A typical driver approach could be the single-spin flip Metropolis method for a spin system, whereas generalized-ensemble methods include parallel and simulated tempering, multi-canonical and flat-histogram methods, population annealing, and certain machine-learning techniques. It was recognized that work is required to advance the toolset on both fronts, by improving the driver methods, for instance through making them irreversible, but also by finding novel ways of beating the slowing down observed for systems with complex free-energy landscapes through improved meta-algorithms, for example adaptive variants of population simulation methods. A somewhat sobering observation that was discussed is that of the more or less comparable performance of a range of generalized-ensemble methods for the simulation of the Edwards-Anderson spin-glass model that can be

seen as a paradigmatic example of a hard optimization and simulation problem. It might indicate that no substantially more efficient approaches can be found for such problems, but this view was not shared by all participants, and so the challenge to find more efficient approaches for such problems remains. A number of promising applications of machine learning methods for the improvement of Monte Carlo simulations was discussed and the participants agreed that it will be very interesting to see the potential of this new set of methods for improving Monte Carlo methodology unfold. At the same time, it was discussed that the heightened interest with which such work is currently met due to the machine learning "hype" also produces a significant body of work of questionable quality.

## 3 Community needs

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The community naturally has significant needs in computational resources, but as it was also discussed at the meeting only rather few projects are conducted at large scale national supercomputing centres. Instead, most groups rely on local resources available at the group, department or university levels which are more suitable for the typical use case of a rather large variety of different projects with small to medium-size computational requirements. Cluster machines with accelerator devices such as GPUs can be useful in this setting for certain projects with larger computational appetite. The present workshop was mostly attended by statistical physicists, but it emerged in the discussions and through the participation of a few researchers with a more mathematical or statistical background, that there are many parallel developments to the ones discussed here in the field of statistics, where Monte Carlo methods are used for hypothesis tests, Bayesian inference and many other applications. Due to the differences in notation, typical examples used, but also the different meetings attended, there is relatively little exchange between these communities. CECAM could help in bridging this gap by facilitating workshops at the interface of these fields (perhaps also including the data science and machine learning community), a task that promises rich rewards in terms of synergies and cross-fertilisation.

## 4 Funding

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Most research presented and discussed at the conference was supported by grants awarded through national funding bodies such as DFG, CNRS, NSF or EPSRC. Apart from bi-national agreements between research councils that would allow joint work between partners in two countries, the main funding source for joint proposals is through the Horizon 2020 scheme of the European Commission (for the majority of attendees hailing from core or associated countries of the European Union). As most of the work discussed was of a rather fundamental nature, this mostly only includes the Marie-Sklodowska-Curie actions and, in principle, synergy grants from the European Research Council (ERC). A subset of the researchers attending the workshop were participants of the FP7 network DIONICOS of the European Commission in the International Research Staff Exchange Scheme (IRSES, now superseded by the RISE scheme) extending from 2014-18. Similar initiatives, also including Innovative Training Networks (ITNs) on the subject would be interesting vehicles for future collaboration.

## 5 Will these developments bring societal benefits?

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The research discussed at the meeting was of a fundamental nature and hence not immediately relevant for commercial applications. Nevertheless, Monte Carlo methods are used in a wide range of contexts, including optimization problems encountered in industry, in statistical inference with manifold applications in actuarial statistics and data science, in the simulation of biopolymers with manifold applications in the life sciences, or for simulations in materials science. The research discussed at the workshop can be seen as the toolbox that is required for the practitioners working on an extremely wide range of practical problems.

## 6 Participant list

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# Active matter and artificial intelligence

Location: CECAM-HQ-EPFL, Lausanne, Switzerland

Webpage: <https://www.cecama.org/workshop-details/117>

Dates: Sep 30, 2019 - Oct 02, 2019

## 1 State of the art

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Initial reports on the use of machine learning in the field of active matter research have been the starting point of this workshop. The main motivation for this combination arose naturally from the fact that many of the mathematical abstractions in machine learning have been conceived from the ability of living systems to evolve and adapt to complex environmental situations with the help of activity, signaling and signal processing. It is thus quite natural that machine learning, i.e., mathematical algorithms which coarse grain the complex biochemical to simple strategies, have just started to be applied to the field of active matter. Examples of such combinations of machine learning and active matter at the time of the application for that workshop have successfully tackled the soaring of birds in turbulent thermal air flows as well as the motion of artificial microswimmers in complex flow patterns and in collective field taxis. Optimization techniques based on machine learning have been applied to find arguments for specific behaviors of individuals in animal swarms or flocks and first approaches to data analysis have appeared. At the same time with these naturally appearing examples, the complexity of the strongly correlated soft matter systems and their far from equilibrium nature, which often invalidate the approaches of thermodynamic equilibrium approaches causing substantial and fundamental challenges for mechanistic models seems to require new approaches for analyzing data. The strength of neural networks to classify and find modeling complex data sets is currently starting to enter the field of active and soft matter research. The understanding of emergent living active matter based on artificial active matter is thus converging scientific fields such as physics, biology, behavioral science, machine learning and mathematics. The workshop targeted in particular the following questions:

- How can fundamental biological questions be addressed by artificial active matter?
- How can signaling and feedback be introduced into artificial active matter?
- Should artificial active matter be interfaced with machine learning?
- How can machine learning contribute to the understanding of biological systems?
- What is the role of feedback and delay in noisy environments for biological function?
- What are relevant physical/mathematical descriptions of active matter systems?

## 2 Major outcomes

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The field of applying algorithms of machine learning (ML) to active matter systems is quiet new uniting two fields which have already developed their own communities. The workshop has demonstrated that first successful steps in the application of ML are taken. Some of the outcomes of this workshop have been incorporated into a review on the field by some of the organizers which is currently under revision. The list below represents the major topics during the discussion in the workshop and identifies the current lines of work in this field.

- Search Strategies – Search and navigation strategies are a fundamental problem of living active matter, for example, for the purpose of foraging. The major part of the discussion on navigation strategies in different complex environments was based on

the technique of reinforcement learning (RL), which is a tool to enforce optimal strategies based by a reward-based learning. Numerical studies of navigation strategies in turbulent flow fields have been presented as well studies of optimal pathfinding in potential energy landscapes. In all of the cases the RL delivers robust solutions to these navigation problems even under perturbations.

- Collective Behavior – Of particular interest is also the emergence of collective behavior in animal systems. Animal systems show swarming or flocking behavior based on sensory information like vision and based on specific goals, i.e., the prevention of predation. While such behaviors are already "hard-coded" into models, the presented studies addressed the problem of how such behavioral rules may have formed. Multi-agent reinforcement learning modeling has been explored to develop Vicsek like swarming. Also the collective motion of locust has been studied with reinforcement learning.
- Data Analysis – One of the challenging fields not only of active matter but also in soft matter in general is the analysis of experimental data. In this field various promising approaches have been discussed. The use of random forest and recurrent neuronal networks (RNNs) for the classification and quantification of anomalous diffusion data has been suggested. Similarly RNNs have been applied to characterize force fields from experimental data. Various approaches to the analysis of optical microscopy images and image series for the classification and tracking of active matter using convolutional neuronal networks have been presented. Strategies for analyzing large scale information on bacterial swarming to understand the relation between genetic code and macroscopic behavior has been a highlight of the workshop.
- Embodied Intelligence – A new area in the field of active matter and artificial intelligence that has been introduced to the discussion is the field of embodied intelligence, i.e., the amount of information that has been incorporated in the design of active elements to support a complementary amount of intelligence in the control. Besides the presentations on the application of machine learning to active matter several lectures have focused on theoretical descriptions of active matter as well as on the experimental control of active matter. Both of them are important to pin down collective behavior with physical models and to provide the ability to make use of active matter in applications.
- Active Matter Theory – Lattice Boltzmann approaches to explain the collective behavior for interacting active particles have been presented to explain the appearance of bacterial turbulence. Also steps to develop a generalized response theory for active matter have been discussed.
- Active Matter Control – Techniques for the control of synthetic active matter are an intermediate step on the way to develop micrometer-sized active materials with sensing and processing capabilities. Current experimental approaches to control individual micrometer-sized active particles in larger ensembles are still rare and all based on optical techniques. The application of specific interaction rules is pursued to provide model systems for signal-based cohesion. First experimental incorporation of reinforcement learning into active particles has also been demonstrated. Besides these specific topics, there has been also a critical discussion on the machine learning approach itself providing a "black box" capable of modeling data with the possibility of over-interpreting its capabilities and results.

## 3 Community needs

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From the discussion of the presentations and posters the following general needs emerged:

- Training Data – Especially in the field of data analysis from microscopic images and time series, the generation of training data sets can be based on model calculations. Complete microscopic images may be simulated to train neural networks. Such training data and algorithms and computer codes provide the ground truth of model generation can be made available to the community within a network of researchers.
- Experimental Data – The second important point is the availability of experimental data that is provided to evaluate the performance of newly developed machine learning algorithms. Similar to the availability of training data for machine learning purposes in this field a platform for experimental data will be beneficial.
- Algorithmic Competition – The same tasks may be tackled with different machine learning algorithms. For example, data analysis of anomalous diffusion data has been demonstrated with random forests and recurrent neuronal networks. To compare the performance of such algorithms standardized testing scenarios are needed.
- Algorithm Availability – One of the major benefits for the community of active matter research will be the free availability of frameworks that may be adapted. A quick growth of the field of machine learning applied to active matter may require a specific database or publication type to collect the approaches specific for this field.
- Computational Power – Most of the computational power required for training supervised approaches relies on the use of graphical processing units. The availability of GPU clusters with general access also to training data seems to be a key issue for the success of such approaches also in the field of soft matter research.

## 4 Funding

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Typical funding sources of the contributors are the national funding agencies as well as the European Research Council. The possibility of joint research proposals has been discussed in the concluding round table discussion with all contributors of the workshop. All of the contributors welcomed the possibility to engage a collaborative network to join forces, highlighting the need for an exchange of students, visiting researchers and regular workshops and meetings. In particular two main possibilities have been suggested: 1) Marie Skłodowska-Curie Training Networks One of the specific joint effort that has been discussed is a Marie Skłodowska-Curie International Training Network, that would allow the participating researchers an exchange between different institutions to establish platforms for the exchange of data, algorithms and know how. 2) COST action A second possibility is the application for a COST action as funded by the European Union. The COST action would also allow for short term exchange of researchers as well as workshops. The required steps for such networks are currently evaluated by a smaller group of the participants of the workshop.

## 5 Will these developments bring societal benefits?

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Overall, the approach of introducing artificial intelligence into active matter research delivers a number of possible ways to pursue that could bring societal benefits. The understanding of navigation strategies in turbulent flows, which was initially motivated by biological questions relating to plankton may be, for example, beneficial for autonomous sensorial networks for monitoring oceanic flows. Exploring huge datasets of bacterial growth may yield substantial new information on drug resistance, biofilm growth and prevention. Exploring communication and synchronization in swarms of animals may provide hints for autonomous driving approaches for future traffic control. The study of control schemes of artificial active matter may finally also pave the way to the development of autonomous microrobots which will find applications in personal medicine, drug delivery or nano-surgery.

## 6 Participant list

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# Modeling phase separation in health and disease: from nano- to meso-scale

Location: CECAM-FR-GSO

Webpage: <https://www.cecam.org/workshop-details/141>

Dates: Sep 30, 2019 - Oct 02, 2019

## 1 State of the art

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The goal of the workshop was to review the recent developments in the field of numerical and analytical models in the physics of life, more precisely in systems where phase separation phenomena are at stake. Phase separation is exploited by biology to organize a number of cellular processes. Examples include heterochromatin in transcription regulation, ribonucleoprotein granules for spatial control of translation in neurons, and phase separation as a key mechanism for the functional organization of the cell membrane. Phase transitions also play important roles in the development of human disease. For instance, RNA phase transition may be involved in severe neurological conditions such as Huntington disease and muscular dystrophy. Liquid-liquid phase separation promotes the aggregation of the intrinsically disordered protein tau to amyloid fibers, which may play a crucial role in the pathogenesis of Alzheimer's disease. Cataract ensues from the formation of insoluble protein aggregates that affect the transparency of the eye lens.

Numerical and analytical approaches have known a recent spectacular development in this context, in order to understand the mechanism and consequences of phase separation in cellular biology. Confronting the models with experiments is of course of primary importance to validate these approaches. Top-experimentalists, leader in their domain, also participated to the workshop to help identifying the key-issues and to keep a critical look on the models.

As phase separation is a ubiquitous mechanism visible from the nucleus to the cell membrane, the workshop gathered researchers working on these different subsystems to generate fruitful discussions and create bridges in-between fields of research.

## 2 Major outcomes

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The CECAM workshop gathered about fifty persons coming from around the world (mainly Europe, plus USA and Canada) and lasted three days, from September 30 to October 2, 2019. These people were scientists with complementary skills (from experimentalists to theorists), from different scientific communities (physicists, biophysicists, high performance programming), senior researchers as well as younger scientists. By promoting debates on the recent progress in these related topics, it has favored bridges between European researchers and world-wide actors in this field.

The talks were divided in three main themes: 1/ Cytoplasm and proteins, 2/ Membrane and lipids, 3/ Nucleus and nucleic acids. Several participants told us that they were very glad to have participated to this workshop because it enabled them to realize that their own field of research had strong connections with those of other speakers, what they had not necessarily anticipated before the meeting. To us, having brought together people from different but connected fields is the main success of this workshop. The discussions and debates were

lively and fruitful, and the last discussion, presented by us as a serious game, was also very instructive.

All in all, the workshop allowed researchers from different subfields of biophysics and biological physics to realize that phase separation phenomena are ubiquitous in cell biology, as is exploited by cells to organize a number of subcellular processes. In prokaryotic cells lacking the internal organelles of eukaryotic ones, a certain degree of organization must prevail and phase separation is evoked as a leading mechanism to avoid random localization within the cytoplasm. In this context, intrinsically disordered proteins, another emerging topic of research, are believed to play a key role. Phase separation phenomena also plays important roles in the development of some human diseases. Sharing these different viewpoints will undoubtedly lead to useful cross-fertilization between these different fields of research.

## 3 Community needs

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This CECAM workshop mainly gathered fundamental research scientists, whose needs essentially rely on high-performance computing resources at this stage. Accessible regional, national or European HPC facilities remain fundamental in this highly-demanding and competitive context. The complex systems under study generally involve many molecular species, some of them of great complexity (e.g., intrinsically disordered proteins (IDP), cell membranes, chromatin), and their simulation requires to consider hundreds of thousands of atoms. One part of solution dwells in using coarse-grained approaches (e.g., MARTINI force-field) or mesoscopic models, enabling to reach larger system sizes and longer simulation time-scales. Consequently, an effort must also be made in the development of these alternative approaches, notably by expanding the validity of coarse-grained force-fields to new classes of objects such as IDP or nucleic acids.

Of course, this emerging community must go on being structured, and national, European and international event organization remains critical to this respect. CECAM workshops are an excellent format to gather a limited number of specialists of such an emerging community, and should be encouraged in a near future.

## 4 Funding

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As explained above, the funding of HPC facilities by regions, states and Europe remains vital to ensure the competitiveness of our research community, in Biophysics and Soft Condensed Matter in general, and in the emerging field of phase separation in cells in particular. Given that fundamental research groups generally have limited financial resources, the fact that access to HPC is freely available must be fervently defended. In this respect, CECAM is an important actor.

As this community is not presently sufficiently structured, we did not discuss the possibility of ambitious large-scale research proposals but this should probably be envisaged in a near-future.

## 5 Will these developments bring societal benefits?

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As discussed above, phase separation of cellular material into mesoscopic or macroscopic liquid-like droplets has recently emerged as a fundamental organizing principle in cells, notably in prokaryotic ones. However, eukaryotic cells are also concerned, for example in the nucleus, where diverse objects such as the nucleolus, nuclear speckle or Cajal bodies are the results of phase separation. Genetic regulation is thus probably deeply related to the underlying physical organization of the genetic material, and this is an emerging and promising field of research, with enormous potential applications in terms of deciphering molecular mechanisms involved in cancer or bacterial infections, with the ultimate goal of developing new therapeutic strategies.

The phase separation processes accompanying some well-known diseases such as Alzheimer disease (amyloid plaques accumulation in neurons) or cataract (clusters of aquaporins in the eye lens) is also an indication that new therapies could ensue from a better understanding of the molecular mechanisms at play in this context.

These non-exhaustive examples show that strong societal benefits can be expected in the mid-term from these fundamental research topics.

## 6 Participant list

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# Electron-phonon coupling: Computational methods for electronic transport in nanostructures and in bulk materials

Location: CECAM-Lugano, Lugano, Switzerland

Webpage: <https://www.cecamlugano.org/workshop-details/110>

Dates: Oct 14, 2019 - Oct 16, 2019

## 1 State of the art

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Real-life performance of semiconductors and metals, whether it be in one, two, or three dimensions, is often limited by carrier scattering by phonons. The mobility of charge carriers is a key parameter in the semiconductor industry to describe the electrical performance and the movement under applied electric fields. The traditional approach to calculate phonon-limited mobility is based on the Boltzmann transport equation in combination with the effective mass approximation and empirical deformation potentials. In recent years predictive parameter-free mobility calculations have been carried out at the density functional theory (DFT) level for the electron-phonon coupling (EPC). EPC may also lead to a Bose-Einstein condensation of electrons near the Fermi surface as Cooper pairs, resulting in conventional superconductivity at sufficiently low temperatures or high pressures. Also, here DFT calculations of EPC have explained the origin of superconductivity in a range of materials and provided quantitative estimates for the critical temperature using Migdal-Eliashberg theory. A recent example includes first-principles theory that revealed how high-pressure hydrogen sulfide is a strongly anharmonic superconductor. In a different context, the introduction of Inelastic Electron Tunneling Spectroscopy has opened up the possibility to study adsorbates and molecular junctions and to characterize inelastic scattering against vibrations down to the single-molecule limit. Ab-initio approaches based on DFT and nonequilibrium Green's functions (NEGF) have been developed to describe the EPC in such nanoscale junctions and to explain the inelastic transport characteristics. Despite the common origin of EPC in these three distinct phenomena (phonon-limited resistivity, phonon-mediated superconductivity and IETS) the systems have traditionally been considered separately by different researchers and with different ab initio methods. In this workshop we gathered researchers interested in such EPC physics from these three different communities using very different approaches, codes, and methods. This provided fruitful discussions where new ideas were inspired by the crossbreeding of the different communities.

## 2 Major outcomes

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The workshop contributed with talks and discussions within each of the three areas dealing with electron-phonon coupling. In addition, discussions on spin-relaxation and photocarrier thermalization arose which could well be considered as additional areas for discussion in future workshops. Some key advances that were discussed at the workshop included:

- Recent advances in ab initio IETS theory including the extension to 2D delocalized phonons at interfaces, second-order processes of IETS at interfaces, and Monte Carlo approaches together with quantum transport calculations compared with the Boltzmann transport equation.
- The importance of anharmonicity in the contexts of phase transitions, charge-density waves, and stabilization of crystal structures was discussed. The role of the quantum nature of light-element atoms (in particular hydrogen) was exemplified for the new class of the high-pressure, high-temperature superconductors H<sub>3</sub>S and RhH<sub>11</sub>.
- Ab-initio modelling of EPC and electron mobility in 3D materials. A high throughput computational search for novel, never synthesized, high-mobility 2D materials was presented and discussed. One of the problems here is about how to deal with the computational complexities of the large number of k/q-points needed for 3D models. New ideas included the change of basis from k-space to symmetrized basis functions which capture most of the EPC on the Fermi-surface without the need for a complete basis. Also, a neural-network strategy for EPC trained with ab-initio data was presented, with perspectives that extrapolation to similar, but larger supercells may be possible. In addition, extension to the Boltzmann approximation was detailed.
- The role of EPC was further described in the relaxation of spin, important for spintronics and quantum computation devices. In addition, for photovoltaic applications the thermalization of the electron-hole excitation and separation of charges is driven by EPC.

## 3 Community needs

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During the workshop dedicated time for discussion in smaller groups was included in the program. The participants were chosen to represent the different subfields present at the workshop and was furthermore changed between days. After initial skepticism, the discussion sessions were appreciated. Summarizing the discussions:

- There is a need for more compatibility between different computational codes. Among the participants, there was widespread use of i) Quantum Espresso, ii) EPW, iii) Siesta, iv) PhonoPy, v) VASP, vi) Wien2K, and (vii) Crystal. For the present workshop the calculated phonon band structures and EPC are all saved in different file formats and there does not exist any test cases or benchmarks which can be used to improve the quality of the research. One of the problems inherent in any benchmark of electron-phonon coupling is the large amount of data needed to describe the mobility using 3D Fermi-surfaces. Recent advances discussed during the workshop where a change of basis from k-points to symmetrized states on the Fermi-surface could perhaps make this easier.
- The existing computational infrastructure and interactions with experimentalists was deemed sufficient as shown during the numerous talks of the workshop.
- The format of the workshop, where different communities within condensed matter physics were invited to participate, was appreciated by the participants. Discussions about a follow-up workshop was encouraged and several participants expressed an interest in organizing a workshop within a couple of years, expanding the participation with additional communities interested in EPC. An existing initiative to set up a related EPC workshop in San Sebastian, Spain, during June 17-19, 2020 was also mentioned.

## 4 Funding

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Funding opportunities and possible applications were not discussed publicly during the meeting. However, the different communities certainly have joint funding and applications for funding within their respective communities. We expect that new collaborations will sprout from this workshop since many of the researchers met for the first time. We expect informal collaboration will start followed by more formal collaboration including joint funding applications. During the discussions in smaller groups a commonly expressed opinion was that the funding agencies often value applications of the computational methods to novel materials over the development of new methods and maintenance of existing computer codes. Also, it is hard to get funding to perform code benchmarking and establish reference data for the community.

## 5 Will these developments bring societal benefits?

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The understanding of electron-phonon coupling is important in a number of technologically advanced materials. The main materials discussed during the workshop include i) molecular materials designed for solar cells where the energy relaxation by phonons allows the photocurrent to be harvested, ii) two dimensional materials that may replace traditional semiconductors in electronics devices where the electron-phonon coupling determine the current carrying abilities of the devices, iii) the search for new superconductors where the electron-phonon coupling is responsible for the formation of the superconductivity, and iv) the use of high throughput calculations of physical properties searching through possible new materials never before synthesized experimentally. These possibilities for new technologies in sustainable energy production, novel semiconductor materials, and energy savings with superconductors have a large potential to affect society not only for economic growth but also towards more sustainable energy production.

## 6 Participant list

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### Organizers

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# WaterEurope: multiscale simulations and coarse-grained models for water and aqueous systems

Location: CECAM-HQ-EPFL, Lausanne, Switzerland

Webpage: <https://www.cecam.org/workshop-details/86>

Dates: Oct 21, 2019 - Oct 23, 2019

## 1 State of the art

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The outstanding complexity of water's behavior makes it hard to predict its properties. The phase diagram of water itself is constantly enriched with newly discovered phases every few years. The water phase diagram is extremely rich, with more than 16 crystals and several amorphous ices, with an unexpected balance between long-range correlations and local order. Its complex interactions have suggested the possibility of more than one liquid phase in bulk or in ionic solutions, as well as hexatic phases in confinement. Fully agreement of numerical results with experimental properties is still an aim of the computational models, that are continuously refined at all the modeling scales to this purpose.

Water constitutes a natural environment for biological molecules: the water activity is critical in phenomena such as in stabilizing cell membranes or RNA-protein complexes, as well as in determining protein structures, protein-protein interactions and proteins-folding rates. The adoption of a multi-scale approach when studying biological-relevant problems is particularly suitable because it provides the theoretical framework to study a variety of aspects that occur at different length- and timescales.

To discuss the state of the art in the field of water research and related areas, we invited senior experts active in the field from different branches of science, with the goal of providing an exciting and stimulating opportunity for theoretical and computational scientists to interchange viewpoints, favoring the contamination of new ideas and the proliferation of new collaborations. Furthermore, the vibrant participation of junior researchers and even newcomers to the field added enthusiasm to our efforts.

## 2 Major outcomes

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1-Many-body effects in water. J. Crain discussed many-body correlations within an electronically coarse-grained model for water, and T. Head-Gordon their relevance in catching thermal and structural effects in many-body force fields for water: Reaching spectroscopic accuracy. T. Kühne presented how Instantaneous asymmetry in aqueous systems correlate with the HB covalency, and T. Urbic hiw to recover the strength of Hbs and anomalous properties of water by simple model. F. Leoni and D. Liberati presented works based on machine learning.

How to model the cooperativity of water? Which strategies shall we adopt for the parameterization of polarizable force fields?

2-Toward multiscale modeling for bio-systems. A. De Simone discussed biophysics of waters at protein surfaces and proteins at water interfaces, and F. Sterpone toward microscopic simulations of crowded cell-like environments. V. Tozzini reviewed Implicit water treatments in molecular dynamics with low-resolution models, and J. Jin presented a new coarse-grained model for water with enhanced structural and thermodynamic properties.

Are the properties of water at extreme conditions important for biological systems? How to coarse grain water without losing properties? Do we need a new model for each problem, or a rational and common strategy?

3-Phase diagram, extreme conditions and large-scale simulations. R. Prosimiti showed the ability of different potentials, based on a hierarchy of approximations, in modeling interactions for determining molecular properties of aqueous systems. P. Gallo discussed atomistic models in dynamic crossovers of relaxation times and structure of trehalose hydration water upon supercooling. J. Russo reviewed what we learned so far from coarse-grained water models and L. Coronas presented a phase transition analysis between low density and high density nanoconfined water.

How to improve interaction potentials in describing the variety of water phases? Can we help in designing how to explore currently unavailable phases? Is a universal model that accurately reproduces water properties from gas to solid realistic?

4-Aqueous solutions and interfaces. A. Michaelides introduced interfacial water: from atmospheric ice nucleation to nano-confinement. M. Berkowitz discussed when to coarse-grain or not to coarse-grain with examples about aqueous solutions of electrolytes and water under shock waves. J. Faraudo presented MD simulations of water at nanomaterials surfaces, H. Lu GTP and oncogenic protein binding at cell membrane, P. Honegger water encapsulated in cellular mimics, A. Pak Systematic Representation Of The Hydration Layer For Highly-Coarse-Grained Solvent-Free Lipid Models. I. Skarmoutsos focused on understanding the interactions of water with nanoporous materials to develop environmental applications, E. Guardia on the force matching approach to multiscale simulations: ion-water force fields from Car-Parrinello data, A. Henao, J. Lan and J. Pan on different hydrated interfaces.

How can we explain specific ion effects? Is it necessary to use polarizable force fields? Are there general effects at interfaces or each interface is a new challenge?

5-Large bio-systems simulations. V. Lobaskin discussed multiscale modelling of bionano interface, H. Lopez short-time diffusive dynamics of proteins in a naturally crowded environment, G. Franzese Multiscale Approach For Hydrated Systems: Perspectives For Cancer Nanotheranostic, V. Bianco including water in large scale simulations for protein mixtures. I. Pagonabarraga showed how to account for hydrodynamics effects in collective response and emergent morphologies in swimmer suspensions.

How to coarse grain water keeping consistency with experiments? Are non-transferable models, inspired to colloidal science, a reasonable solution? How can we account for hydrodynamic effects in biological systems?

## 3 Community needs

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Despite its simple chemical formula, water exhibits complexities that spread on a wide range of spatio-temporal scales: from the quantum nature of electrons and hydrogen bonds to the macromolecular folding of proteins and the stabilization of cell membranes. Hence, different methodologies have been developed both from theoretical and experimental standpoints. Ab initio methods, electronically coarse-grained models, classical molecular dynamics, coarse-grained macromolecular models, and continuum theories including hydrodynamic effects are all techniques for which there exist well-established simulation codes which are constantly improved. The need for HPC resources is systematically increasing, with the goal of pushing the limits of each technique to larger time and length scales (from hundreds of ps and few hundreds of water molecules with ab initio to hundreds of minutes and millions of macromolecules with coarse-grained models).

Europe has a remarkable heritage in the study of water properties, with research groups leading the world scene. CECAM has contributed to this environment by organizing several successful workshops on the modeling of water under different conditions, often focusing on specific length and time scales associated with particular problems. Nevertheless, there are still many challenges related to water, as emphasized by the high number of open questions that emerged in our workshop. The large need for improving our ability to approach problems that are biologically and technologically relevant and the intrinsically multidisciplinary nature of these problems strongly recommends the organization of a series of dedicated CECAM workshops. This would help in strengthening the networking among different communities interested in hydrated systems, including experimentalists and theoreticians in physics, chemistry, biology, geology, climatology, space science, nanoscience, engineering, food science, pharmacology, and medicine.

## 4 Funding

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Typical funding channels are national and international calls for basic and applied research. Blue-sky funding schemes, such as ERC grants, and more application-oriented EU calls, as in the Horizon 2020 framework, are typical examples. Within the coming Horizon Europe, the planned 7-year European Union scientific research initiative meant to succeed the current Horizon 2020 program, new funding sources will likely be present in each of the pillars: Pillar I – Excellent Science (including ERC, MSCA, and research infrastructures as HPC and exascale resources deployed under the European Data Infrastructure); Pillar II – Global Challenges and European Industrial Competitiveness; Pillar III – Innovative Europe; and the transversal activities aimed at widening participation and strengthening the European Research Area. The possibility of joint research proposals was not explicitly discussed during the meeting. However, several collaborations have been encouraged and started.

## 5 Will these developments bring societal benefits?

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Water is the basic ingredient of our life and is an essential part of many industrial processes and technological applications. The study of its properties has many potential societal benefits.



For example, understanding how water interacts with interfaces and ions has a large interest in water purification and sanitation. Addressing water quality is part of the objectives of the Horizon Europe implementation, with which Europe wants to take the lead on the *transition to sustainability*, as strongly demanded by society. As the populations of the world continue to expand, access to drinkable water becomes a global issue. Although elected a *fundamental and universal human right* by the United Nations General Assembly in 2010, its scarcity is making water the world's most crucial commodity, now planned to be put on the board for investors alongside other raw materials like crude oil, soybeans, and copper [December 2020]. Water research centrality in climate change is also evident, e.g., considering the relevance of better basic comprehension of the water molecular mechanisms in ice nucleation in clouds and seas. Hydration and ice formation play a fundamental role in food security, bio-based industries, environmental degradation, and pollution. Gaining a proper understanding of how water stabilizes biological systems and how water interacts with membranes or drugs, all topics treated in our workshop, is a fundamental step for a proper, rational design of new drugs devoted, e.g., to find new cancer treatments, or to defeat antibiotic resistance—one of the biggest threats to global health, according to the World Health Organization. The 173 occurrences of the word "water" in the 142 pages of the EC document describing the strategic plan for Horizon Europe quantify the centrality of the research topic of the workshop in the future societal transformations.

## 6 Participant list

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### Organizers

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#### **Franzese, Giancarlo**

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# Protein simulations - current state of the art

Location: CECAM-ISR

Webpage: <https://www.cecarn.org/workshop-details/153>

Dates: Oct 22, 2019 - Oct 24, 2019

## 1 State of the art

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The workshop was meant to follow on the technological and theoretical developments that enable studies of protein dynamics at previously unattainable resolution from experiments. At the same time, advances in hardware and coarse grained forcefields and parameters made calculations of large macromolecular complexes and even cells possible, as demonstrated by talks that were given in the conference. The following topics were presented:

- Current state-of-the-art in terms of force-fields, timescales and enhanced sampling methods
- Modelling of cofactors and metal ions
- Estimation of anisotropic displacement factors and configuration entropies from simulations
- Where we as a community should put more effort in order to advance the field in the next three years and beyond (improved classical forcefields, polarisable forcefields, coarse grained simulations, enhanced sampling methods, or something else).

At the discussions additional subjects were addressed, owing to the demand from the participants, as described below.

## 2 Major outcomes

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The following topics were debated during a discussion session:

1. Should we try to perform very long all atom simulations?
2. How can we move towards a more universal forcefield?
3. How to study the effect of solutes and how important they are.
4. Quantification of the entropy and its components.
5. Handling large protein complexes.
6. Coarse grain (CG) models and how they correspond to atomistic ones.

With respect to the scale of the simulations, the participants came to the conclusion that running longer all-atom simulations is not the best way to move the field forwards. Rather, all-atom simulations should be repeated many times for better sampling, and CG models can be used to achieve longer simulation times. The same was judged to be usually true also for system sizes - the participants suggested that large, multiprotein complexes should be studied with CG models, since it is anyway difficult (and often not useful) to analyse them atom-by-atom. When it comes to forcefields, it has been pointed out that significant improvements were achieved in recent years in describing e.g. nucleic acids and sugars. Polarizable forcefield are still not widely used in the community. Multivalent ions still pose a challenge to forcefields as are protons and protonation states, although constant pH simulations are being gradually more common. The audience did not seem supportive of flexible bond models. Solute effects were recognised as important, and it is expected that studies that quantify them will be published

more often. The same is true for entropic effects - efforts are made to calculate them more accurately, but experimental references are still lagging. The correspondence between CG and all atom models was lively debated. Some speakers would like to see a more uniform approach, where CG-models are transformed to all-atom models and vice versa. Others argued that as CG models provide insights on different timescales (and often also different sizes), we must use the right tool for the problem at hand rather than moving back-and-forth between models. Overall, many of the workshop participants took part in the discussion and told us that they found it very useful. Our plan is to summarise it in the Editorial for a special issue of the Israel Journal of Chemistry devoted to the workshop.

## 3 Community needs

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Existing codes work very well for almost all applications that were presented, with few of the participants actively developing such codes (for CG simulations of different scales and calculation of entropies) and many others using them successfully. HPC resources can be the bottle neck in some cases, but with the development of better hardware and software this was not seen as a stumbling block. There is a need for better coupling between theory and experiment. Few of the presenters carry out experiments and could explain them and also follow up on the simulations, but simulation-inspired experiments are yet to become routine. The participation of experimentally-oriented scientists was judged by the participants to be beneficial. Feedback to the organisers was highly positive. A series of similar CECAM workshops should definitely be considered to follow up on the development of the field. It would be interesting to see, for example, if polarisable forcefields and constant pH simulations will be more routinely employed in the future.

## 4 Funding

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When it comes to funding of the presented research, most have been supported by national agencies and designated EU funding programs. Possibilities for funding of joint projects are therefore limited. A problem that was discussed, albeit without the proposition of a decent solution was the reluctance of funding agencies, including some general science funding agencies to support studies that are purely theoretical without clear applications as yet. That is not to say that such studies are not being supported, but some participants found that it has been difficult to be awarded such funding. One of the (mostly) experimental scientists in the audience has specifically mentioned a reliance on theoretical methods developed by others to supplement experimental studies, as it is difficult to sustain research groups that are expert in both aspects. This might be conveyed to granting agencies that are examining inputs from the scientific communities. Many of the presentations relied on the use of high performance computing units (HPCU). Another source of computer time was the private company D. E. Shaw - several researchers reported that they applied to, and received, the right to use the company's proprietary computing system, Anton.

## 5 Will these developments bring societal benefits?

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The research topics that were presented and discussed have many potential societal benefits. Some of the proteins that were studied are drug targets, or play important roles in diseases. In addition, studies reported on issues that are of strong interest to the industry such as taste recognition. Improved ways to simulate proteins could also benefit the field of enzymatic design, which is very important for the biotech industries. Few participants work at, or collaborate with the industry. They were interested in many of the topics that were raised for their application in biotech. It can be anticipated that the industry can make use of future developments in the field, especially those that are open source and fairly straightforward to use. Sustainability is also a potential societal benefit since enzymatic catalysis is greener than traditional chemical catalysis. It can thus be anticipated that calls aiming at green chemistry can appeal also to scientists that deal with protein simulations.

## 6 Participant list

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### Organizers

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**Friedman, Ran**

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# Progresses and challenges in modeling activated phenomena: from machine-learned energy surface sampling to multi-scaling approaches

Location: CNRS-LAAS and University of Toulouse Paul Sabatier

Webpage: <https://www.cecarn.org/workshop-details/105>

Dates: Nov 12, 2019 - Nov 15, 2019

## 1 State of the art

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There are no changes with respect to the submitted proposal. The workshop has been fully compliant with the proposal state-of-the-art, goals and expectations. In the following the submitted state-of-the-art. Modelling activated phenomena is one of the most active trans-disciplinary fields in computational material science. Its applications and outcomes impact domains as broad as bio-chemistry and chemistry, surface and interface physics and materials under manufacturing and working conditions. Three inter-connected computational aspects determine our modelling capacity in this context: the accuracy of the energy/force model, the efficiency in identifying reaction paths and the multi-scaling, i.e. the ability to reach meaningful length and time scales. Nowadays approaches such as Density Functional Theory, in all its flavours, have proven their accuracy. However, their success often falls short when the model system exceeds few hundred atoms or the target time scale is larger than a few picoseconds. The computational price of accurate energy/forces models becomes rapidly unaffordable when the understanding of an activated phenomenon requires a careful exploration of the energy landscape together with the identification of the relevant saddle points. Dynamically speaking, when a system is in a local minimum and the kinetic energy is lower than the surrounding activation barriers, the system spends most of its time exploring a relatively narrow region around the minimum. Indeed, activated phenomena are also called rare events. Even in the case of overcoming one of the barriers, because of the instability of the system at the saddle point, the time spend at it is negligible and, as such, the evolution from one minimum to another results in a “jump”. The early attempts to overcome these drawbacks were based on Lagrange formulation of constrained dynamics, as in Drag methods. One of the most successful approach has been, and still is, the Nudged Elastic Band (NEB) method developed by Jonsson et al in the late 90’s, which, alone and/or combined with Object Kinetic Monte Carlo has demonstrate significant breakthroughs. However, the human workload necessary to meaningfully explore energy surfaces with NEB, for feeding eventually an OKMC, is an obstacle to model complex systems. In the 2000s several approaches to reduce the human workload have been developed, such as Metadynamics. The search is, however, usually restricted to a finite number of intuited relevant generalized-coordinates. Fully automatic and blind approaches are also available, as for instance the combination of hessian estimation with KMC. In all cases, due to the very large number of force evaluations needed, the application of automatic or semi-automatic search is mainly restricted to classical force fields interaction models. The recent boom of Machine Learning approaches is opening new perspectives, indeed neural networks have been added to on-the-fly and kMC algorithms for shortcutting part or all the cpu-intensive calculation of energy and forces. But, is the future really that bright? Are these frameworks ready to be exploited as black boxes? In other words: how dependent is the success of such combined approaches on the targeted systems? How general could be

a given software platform? How could the number of training points, near and at the saddle point, be optimise for optimal sampling in Machine Learning perspective?

## 2 Major outcomes

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Multi-scaling needs fast and accurate energy-force-Hessian engines! but “Waiting for faster computers is not an option!”. So, the alternative are Machine-Learned potentials. Everybody agrees that sampling around saddle points is always poor. Is there any hope? There were some suggestions that hot MD could solve the problem: every single barrier might not be right but on average the dynamics should be reasonable. Why not to run a dynamics with something that is linear in the momentum? Maybe this might explore the phase space more richly. There are still many open questions and challenges! Still on the ML side the construction of ML potentials is computationally demanding, non physical functional form (validation is required, but how to?). There is no way to fit both the chemical space and the configurational space! How to deal with long range interactions? There are hardly any reliable comparison of different methods and unbiased comparisons are really difficult. For instance, the literature is full of new proposed methods which have not been tested (or tested only for customised cases). The scaling of the number of descriptions with chemical diversity. How to reduce the number of reference data. How to scale with descriptors (local vs global description)? Most importantly, the ML community, need to start providing error estimate!!!! and benchmarking the results! In a more fundamental view, it is not fully clear what is the theory behind Neural Networks; What functions can we fit? ANY? What are optimal parameters? Well performing NN, are crafted with an architecture that craft very well the problem that they try to solve. Any NN cannot work for everything. How to decide? How to design? (For more details, a full workshop report has been sent to CECAM, and it will be made available by the ML4MS organisers)

## 3 Community needs

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During the round table several interesting questions have emerged, mainly addressed by PhD students and young postdocs. To cite the most discussed ones: i) The access to data is becoming an important drawback. Not only for data sets, i.e. links to data are sometimes provided in the reference papers, but there is no central repositories. With the boom of ML papers, it is difficult to get an idea of available data sets, but even just at the level of the published figures. It is difficult to have access to the files xy that generated a given figure. In the past, it was possible to use some graphical tools to get the points by hand, but with the large amount of data points in figures (some times of the order of several thousands) it is impossible to continue with “hand”. It would be good if, as supplementary material for instance, the data points of the figures were also made accessible. ii) Regarding data, the case of plumed-nest was discussed as an interesting inspiration, that could be maybe applied in ML.

## 4 Funding

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The workshop has been co-financed by CNR-IOM, CNRS-LAAS and CECAM node SNS-SISSA. Two young researchers (a PhD student and a postdoc) working in South-Africa were able to attend the workshop thanks to the support of ASES MANET (<https://eaifr.ictp.it/about/news/asesmanet/>) and ICTP-EAIFR (<https://eaifr.ictp.it/>). At the round table the possibility for COST application has been discussed. And it is an eventuality that the organisers are seriously considering. It has also been discussed to maybe exploit a present COST Action (CA17126), in which the ML4MS organisers and some participants are already involved, to start an “experiment” on data sharing. The combined field multi-scale and machine learning is very heterogeneous. As far as we know, it is the first time that such a “all-together” meeting has been done. Behind clear common interests and complementarity (for more details, a full workshop report has been sent to CECAM, and it will be made available by the ML4MS organisers). Probably some additional meetings and/or workshop would be need to converge on more concrete path ways to gather and share efforts.

## 5 Will these developments bring societal benefits?

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The field is at the moment at the beginning, and still much work has to be devoted to improving the available methods. The successful development of both multi-scaling and machine learned potential could contribute to the bottom-up technology aided design: optimal material for devices and optimisation of production lines. The field also could benefit the development and testing of radiation-tolerant devices fundamental for the near and far future of MARS missions, and in general, space missions.

## 6 Participant list (Partial)

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### Organizers

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**Combe, Nicolas**

CEMES-CNRS, France

**De Gironcoli, Stefano**

Scuola Internazionale Superiore di Studi Avanzati - International School for Advanced Studies, Italy

**Hemeryck, Anne**

LAAS-CNRS, France

**Martin-Samos, Layla**

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**Richard, Nicolas**

CEA, France

**Salles, Nicolas**

CNR-IOM/Democritos National Simulation Center, Italy



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**Alexandre, Tkatchenko** - Univ. of Luxembourg, Luxembourg  
**Csanyi, Gabor** - University of Cambridge, United Kingdom  
**Jörg, Behler** - University of Gottingen, Germany, Germany  
**Mousseau, Normand** - University of Montreal, Canada

# Polarons in the 21st century

Location: CECAM-AT

Webpage: <https://www.cecam.org/workshop-details/125>

Dates: Dec 09, 2019 - Dec 13, 2019

## 1 State of the art

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The polaron concept was introduced by Landau in 1933, in the context of polar or ionic solids. An electron in such a material will deform the lattice around it, and the resulting quasiparticle (electron+lattice deformation) is called a polaron. In recent years, polarons have taken front stage again in many research disciplines. Polaronic effects are now studied in very varied systems outside of the traditional solid state physics setting where they were introduced. Electrons on liquid helium experience a polaronic mass enhancement, where the ripplons take the role of the phonons. In ultracold atomic gases, impurities embedded in a Bose-Einstein condensate interact with the Bogoliubov excitations of the superfluid in a way that maps onto the polaron Hamiltonian. Rydberg atoms allow to study very large polarons, and the rotational degrees of freedom of molecules have given rise to the "angulon" concept. Also in solid state physics, polarons have seen a big increase in interest in the recent few years. New experimental probes (EPR, STM & AFM, ARPES) allow to study various aspects of polaron physics that cannot be probed by absorption spectroscopy. Polarons have been identified in novel materials, such as lead-halide perovskites and titanium dioxide nanograins, where moreover the polaronic effect has been shown to give a major contribution to those materials unique and technologically relevant properties. Breakthroughs in numerical methods, for example combining ab-initio calculations with diagrammatic monte carlo methods have solved outstanding puzzles such as the width of the relaxed excited state peak in polaron response. These recent developments took place in fairly disconnected communities, which we successfully brought together for this workshop.

## 2 Major outcomes

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The main outcome of the workshop is that researchers in the various communities, hitherto unaware of each others work, got to know each other and got information on what is going on in interdisciplinary polaron physics. This has led to several new insights. A prominent example is the question of polaron formation dynamics. When an electron is introduced in the conduction band (by doping or UV irradiation), the polaron takes some time to form the cloud of phonons surrounding it. This was identified from the start as an interesting open problem. It is a question of importance since it can interfere with some probes of polaron physics: if the probe follows the pump too quickly, the polaron is not yet fully formed. If it follows too late, the polaron may already have decayed or scattered through many-body interactions into other states. In the workshop, we learned from researchers in ultracold atomic gases that they are able to study the polaron formation in a very controlled way using the condensates with a tunable interaction strength (adapted experimentally via Feshbach resonances). The first measurements, performed in the context of quantum gases, point the way to a resolution of the polaron formation in solids. The major methods to identify polaron formation using ab-initio methods were reviewed and updates given on the current development. The limitations of methods such as starting from an initial deformation or supercell methods, were outlined. The main problem was identified, and lies in the fact that the entanglement between the electron

and the phonons is very difficult to take into account using a full ab-initio approach. For small polarons these entanglement effects are not very important, but at weaker coupling, for larger polarons, the entanglement turns out to become even dominant. The workshop also revealed the way ahead to solve this problem. The idea is to combining ab-initio calculations of the parameters of model Hamiltonians (electron-phonon couplings, phonon bands, ...) with semi-analytic methods that start from these Hamiltonians and provide a quantum mechanical description capable of fully taking the electron-lattice entanglement into account (such as diagrammatic Monte Carlo).

## 3 Community needs

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Clearly, more networking is needed between the different communities. This workshop has shown that polaron physics is quite interdisciplinary and topical, but it has also revealed that the different communities (quantum gases, solids, numerical development, experiment, analytical/mathematical developments) are not aware of breakthroughs that would benefit each other. Hence the clearest need laid bare by this workshop is more and better communication between these communities. Repeating this workshop in the future will help alleviate this need. On the computational side, there is a need to integrate ab-initio calculations that yield estimates of Hamiltonian parameters, with codes that take these Hamiltonians as input to compute binding energies, optical conductivity and mobility. Moreover, although fully ab-initio computation of small polarons is currently under development, a similar research programme for large polarons, or one bridging the gap between large and small polarons is still in its infancy and requires more resources (in the form of research funding) to mature.

## 4 Funding

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Current funding channel:

- Austria-Flanders bilateral projects (FWF-FWO lead agency projects)
- Programmes of national science agencies Given the interest, it may be useful to set up an international network or address EU funding.

The topic fits in the "Future and Emerging Technologies" area, and in the societal challenge on clean energy. In this workshop it become clear that many materials that are of interest in this field (for solar energy or hydrogen cells) obtain their technologically relevant properties exactly because of the polaronic effect. Further study of polaronic physics will allow to identify new materials that can tackle the challenges outlined in the EU research and innovation programmes.

## 5 Will these developments bring societal benefits?

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The workshop tackles questions of fundamental physics rather than applied physics or technology. Nevertheless, the workshop clearly illustrated the importance of polaronic physics for future technologies enabling sustainability. In solar cell materials, polaronic physics prevents the photo-created electrons and holes from quickly recombining, allowing to harvest

more charge and boost efficiencies. Perovskite solar cells are an example: they reach 20% efficiency (compared to 10-15% for conventional photovoltaic materials) and can be produced at low cost. Polaronic effects underlie this efficiency. Developing tools that quickly and robustly can identify materials with strong polaronic effects are important in the search for new photovoltaic materials. Another example is water splitting by titanium dioxide nanograins. The polaronic effects also here help separate charge carriers and helps to localize them on the surface of the nanograin where they can interact with the water molecules. Finding better materials for water splitting would be a technological breakthrough that underlies a hydrogen economy.

## 6 Participant list

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### Organizers

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**Devreese, Jozef**

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**Franchini, Cesare**

University of Vienna, Austria

**Kresse, Georg Kresse**

University of Vienna, Austria

**Tempere, Jacques**

University Antwerpen, Belgium

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**Arlt, Jan** - Aarhus University (Denmark), Denmark

**Chevy, Frédéric** - LKB Ecole Normale Supérieure, France

**Christof, Woell** - KIT, Germany

**Ciuchi, Sergio** - Dept. of Physical and Chemical Sciences University of L'Aquila, Italy

**Demler, Eugene** - Harvard, United States

**Deskins, N. Aaron** - Worcester Polytechnic Institute, United States

**Di Valentin, Cristiana** - University of Milano- Bicocca, Italy

**Diebold, Ulrike** - Vienna University of Technology, Austria

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**Giamello, Elio** - University of Torino, Italy

**Giustino, Feliciano** - University of Texas, Austin, United States

**Grioni, Marco** - Swiss Federal Institute of Technology Lausanne, Switzerland

**Klimin, Sergei** - University Antwerpen, Belgium

**Kokott, Sebastian** - Fritz-Haber-Institut Berlin, Germany

**Lany, Stephan** - National Renewable Energy Laboratory, Golden, United States

**Lemeshko, Mikhail** - Institute of Science and Technology Austria, Austria

**Mishchenko, Andrey** - RIKEN, Japan

**Reticcioli, Michele** - University of Vienna, Austria

**Schmidt, Richard** - Max Planck Institute of Quantum Optics, Garching, Germany

**Selloni, Annabella** - Princeton University, NJ, United States

**Shluger, Alexander L.** - University College London, United Kingdom

**Van De Walle, Chris** - University of California, Santa Barbara, United States

**Walker, Alison** - University of Bath, United Kingdom

**Zhu, Xiaoyang** - Columbia University, United States

# **E-CAM Workshops 2019**

# Extended Software Development Workshop: Scaling Electronic Structure Applications

Location: CECAM-IRL

Webpage: <https://www.cecam.org/workshop-details/180>

Dates: Jan 07, 2019 - Jan 18, 2019

## 1 State of the art

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The evolutionary pressure on electronic structure software development is greatly increasing, due to the emergence of new paradigms, new kinds of users, new processes, and new tools. The large feature-full codes that were once developed within one field are now undergoing a heavy restructuring to reach much broader communities, including companies and non-scientific users. More and more use cases and workflows are performed by highly-automated frameworks: high-throughput calculations and computational materials design, large data repositories, and multiscale modeling, for instance. At the same time, HPC Centers are paving the way to exascale, with a cascade of effects on how to operate, from computer architectures to application design.

These trends are highly challenging for the electronic structure community. As a result, long-ignored, well-established software engineering good practices are now adopted at an accelerating pace by more and more software projects. With time, this kind of migration is becoming a question of survival, the key for a successful transformation being to allow and preserve an enhanced collaboration between the increasing number of disciplines involved. Significant efforts of integration from code developers are also necessary, since both hardware and software paradigms have to change at once.

Addressing these issues requires coordinated efforts at multiple levels:

- through the creation of open standards and the use of co-design, both for programming and for data
- with a significant leap in documentation policies, helped by tools like Doxygen, Sphinx and ReadTheDocs
- by introducing test-driven development concepts and systematically publishing test suites together with software
- by creating synergies with popular software distribution systems (e.g. EasyBuild, Spack, or MacPorts)
- by disseminating the relevant knowledge and training the community, through the release of demonstrators and giving all stakeholders the opportunity to meet regularly.

## 2 Training provided

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The first two days of the workshop consisted in presentations and discussions that were roughly divided into four main topics:

- current state and future plans for the Electronic Structure Library;
- methodological and software developments in Electronic Structure;
- current and future trends in high-performance computing;
- performance and scalability of ESL libraries/components.

The participants identified the following aspects as the main challenges that the ESL must overcome:

- The ESL needs more involvement from the electronic structure community to become sustainable in the long term.
- Wider integration by community codes, as code developers are still reticent in using code written by others.
- Increase project visibility by better publicizing the work done within the ESL and increasing the outreach activities.
- Improve the scalability, optimization, and hardware awareness of ESL components in order to ease the efforts of electronic structure codes in addressing the various issues related to the current hardware race.
- Engage more with other projects and communities that share similar objectives.

Concerning the methodological and software developments in Electronic Structure, participants presented how the different projects they are working on are handling the changes in HPC architectures and their use of external libraries. This overview was nicely complemented by a dedicate session on HPC, where it was highlighted that hybrid architectures (CPUs+GPUs) are beginning to dominate the market and that there is still quite some uncertainty regarding what technology will come next. As a consequence, developers of electronic structure codes will have to adapt to these changes in computer architectures, but without necessarily being able to drive them. Different strategies to deal with these changes were also presented and discussed.

A full session was dedicated to the performance and scalability of ESL libraries. Developers were given the opportunity to present the libraries, with a focus on the parallelization strategies used, and the corresponding performance and scalability. They were also encouraged to discuss existing bottlenecks and possible ways to overcome them. This information was essential to plan part of the work to be done during the coding sessions.

A hands-on tutorial on profiling parallel applications was organized before the start of the coding sessions. This tutorial was found to be very useful by the participants and will hopefully lead to more collaborations between ESL developers and the MaX and Pop CoEs.

Lectures from this workshop were recorded and stored on E-CAM's online training portal at <https://training.e-cam2020.eu/collection/5c3329f4e4b0f6515d0b995d>

## 3 List of the software development projects

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During the workshop the participants gathered in smaller groups to work on specific projects. ESL Bundle and Demonstrator

A procedure and a detailed timeline to release new versions of the Bundle with updated components was decided and tested. This led to a new release (0.3.1). It was also decided to

do two new major/minor releases every year. New Docker images were generated for this new release to be used in the ESL Continuous Integration system and a new repository of ESL Easyconfig files for the EasyBuild build and installation framework was created. Work was also done in the ESL Demonstrator to complete some of its features.

#### ELSI

Work on GPU support for ELSI and its solvers was started and benchmarks of the MAGMA eigensolvers and for ELPA 1-stage and 2-stage eigensolvers were performed. The SIESTA-ELSI interface was updated. Several updates to the build system and to the solvers included in ELSI.

#### GridXC and Libvdx

Benchmarks for the two libraries were performed, followed by some more in-depth analysis of the FFT performance.

#### ESCDF

The extension of the API for optimal parallel scalability and performance was thoroughly discussed and work on its implementation was started.

## 4 Participant list

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### Organizers

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#### **Artacho, Emilio**

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#### **Blum, Volker**

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#### **Corsetti, Fabiano**

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#### **MacKernan, Donal MacKernan**

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#### **O'Cais, Alan**

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#### **Patterson, Charles**

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#### **Pouillon, Yann**

Simune Atomistics, Spain

#### **Sanvito, Stefano**

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**Bosoni, Emanuele** - CRANN, School of Physics, Trinity College Dublin, Ireland

**Caliste, Damien** - Alternative Energies and Atomic Energy Commission (CEA), France

**de Jong, Wibe** - Lawrence Berkeley National Laboratory, United States

**Elena, Alin** - Daresbury Laboratory, United Kingdom

**Garcia, Alberto** - Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Spain

**Garcia Suarez, Victor Manuel** - University of Oviedo, Spain



**Gimenez, Judit** - Barcelone Supercomputing Centre (BSC), Spain  
**Gonze, Xavier** - UCLouvain, Belgium  
**Hourahine, Benjamin** - University of Strathclyde, United Kingdom  
**Keal, Thomas** - STFC Daresbury Laboratory, United Kingdom  
**Larsen, Ask Hjorth** - Nano-bio Spectroscopy Group and ETSF Scientific Development Centre, Departamento de Fisica de Materiales, Spain  
**Lueders, Martin** - Daresbury Laboratory, United Kingdom  
**Martin-Samos Colomer, Layla** - University of Nova Gorica, Slovenia  
**Ohlmann, Sebastian** - Max Planck Computing and Data Facility, Germany  
**Payne, Mike** - University of Cambridge, United Kingdom  
**Rampp, Markus** - MPCDF, Germany  
**Saxe, Paul** - MolSSI - The Molecular Sciences Software Institute, United States  
**Wylie, Brian** - Juelich Supercomputing Centre (JSC), Germany  
**Yu, Victor** - Duke University, United States

# Extended Software Development Workshop: Topics in Classical MD

Location: CECAM-FR-RA

Webpage: <https://www.cecam.org/workshop-details/84>

Dates: Apr 03, 2019 - Apr 12, 2019

## 1 State of the art

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This workshop was intended to provide both a venue for scientific discussion, and also an opportunity to learn about the best practices in modern software development, so that participating researchers could hone their tools. From the scientific standpoint, classical molecular dynamics is a broad umbrella, and researchers within specific topics of the field often aren't familiar with the state of the art within other topics. The scientific goal of this workshop was to introduce participants to the core science of several topics and to software related to those topics, while also encouraging them to dive more deeply into a topic of their interest by implementing new functionality as an E-CAM "module." Of course, there are more possible topics than could be covered during the workshop. The three we selected were based on topics that proved interesting at the E-CAM State of the Art Workshop held in Vienna, in October 2018. The topics and related software packages we selected were:

- trajectory-based rare event methods, with software package OpenPathSampling
- CV-based rare event methods, with software package PLUMED
- neural network potential energy surfaces, with software package N2P2.

This workshop was also designed to introduce participants to software development best practices. These best practices have evolved over decades of experience in the software industry. They result in more maintainable and robust software, and facilitate rapid development of new approaches. Many research scientists do not use these best practices, and indeed, many are even unaware of them. As a result, scientific innovation is slowed by bad software development practices. This workshop included training about best practices, and participants were encouraged to use these best practices while implementing their modules.

## 2 Major outcomes

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The presentations during the workshop fell into three categories:

1. introductions to best practices in software development;
2. presentations on the theories and software behind the specific topics the workshop focused on; and
3. contributed presentations from participants regarding their own research.

The presentations on software best practices included introductions to: git and related tools (Alan O'Cais); software testing (David Swenson); object oriented programming (Jony Castagna); performance and benchmarking (Emmanuel Quémener); and git/GitHub/GitLab workflows (David Swenson). The presentations on the theories and software for the workshop topics were: trajectory-based rare event methods and OpenPathSampling (David Swenson); neural network potential energy surfaces and N2P2 (Christoph Dellago; Andreas Singraber);

and CV-based rare event methods and PLUMED (Gareth Tribello). Finally, the contributed presentations were:

- "Augmented Harmonic Linear Discriminant analysis" (Faidon Brotzakis)
- "The analysis of chromatin configurations in the nucleus using MD simulation" (Ali Farnudi)
- "Gold nanoparticles as amyloid- $\beta$  fibril inhibitors" (Francesco Tavanti)
- "Progress index-guided sampling: an unsupervised protocol to boost molecular simulations" (Cassiano Langini) Emmanuel Quémener's presentation on performance dealt directly with some topics in HPC.

The approach of the three software packages presented is generally to gain as much performance as possible from the underlying molecular dynamics codes that they integrate with. The workshop had 21 attendees, including the 8 speakers. In addition, Christoph Dellago's presentation was open to public, and had a total of around 50 attendees (including workshop participants). Ten of the workshop attendees opened merge requests for E-CAM modules they intend to contribute. One recurring theme in the discussions was the question of when and how to contribute to community codes, instead of developing your own from scratch.

## 3 List of the software development projects

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The following modules were in development during the ESDW:

- Add "nucleic" keyword to MDTraj atom selection language ([https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge\\_requests/130](https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge_requests/130))
- PLUMED dimensionality reductions ([https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge\\_requests/131](https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge_requests/131))
- Python bindings for "Open Dynamics Engine" ([https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge\\_requests/132](https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge_requests/132))
- Python bindings for PIGS/CAMPARI ([https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge\\_requests/133](https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge_requests/133))
- OpenMM implementation of d-AFED ([https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge\\_requests/134](https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge_requests/134))
- Update N2P2 weights via artificial MD ([https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge\\_requests/135](https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge_requests/135))
- Tools for analyzing training set size dependence in N2P2 ([https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge\\_requests/136](https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge_requests/136))
- Symmetry function parameter generator for N2P2 ([https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge\\_requests/137](https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge_requests/137))
- Gyration tensor for PLUMED ([https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge\\_requests/141](https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge_requests/141))
- Reweighted path ensemble for OPS ([https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge\\_requests/142](https://gitlab.e-cam2020.eu/e-cam/E-CAM-Library/merge_requests/142))

Both of the modules to create Python bindings will benefit from the HPC performance of the underlying codes. In addition, several modules are being designed specifically with performance considerations in mind. Implementing d-AFED for OpenMM is intended to benefit from OpenMM's GPU acceleration. One of the purposes of developing Python bindings for the PIGS routines in CAMPARI is to interface with MD engines in a way that avoids the performance costs that currently come from using disk as an intermediate. We have not discussed the relevance of these modules with industrialists. However, it is reasonable to expect that some may be of interest. For example, the development of Python bindings for

both the PIGS method in CAMPARI and for the ODE engine are intended to bring methods implemented in those software packages to a wider audience by lowering the barrier to entry. The modules related to N2P2 may be useful for the materials science industry. Although these modules are not scheduled for any particular E-CAM deliverable, they would, once completed, be suitable to include in a future deliverable. These modules were selected based on participant interest; i.e., in response to the requests of users.

## 4 Future plans

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We expect that this workshop will be followed by a second face-to-face meeting. Pending confirmation of the availability of participants, we hope to schedule this for late October. Before the follow-up, we hope that the participants will have completely implemented all the functionality of their modules. The purpose of the follow-up will be to help them bring their modules up to the standards of the E-CAM Software Library, and to complete the documentation of their modules so they can be included in the E-CAM Software Library.

## 5 Participant list

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### Organizers

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**Everaers, Ralf**

École Normale Supérieure de Lyon, France

**Swenson, David**

École Normale Supérieure de Lyon, France

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**Brotzakis, Faidon** - University of Cambridge, United Kingdom

**Buchner, Florian** - Vienna University of Technology, Austria

**Carrivain, Pascal** - Centre Blaise Pascal, ENS Lyon, France

**Castagna, Jony** - STFC Daresbury Laboratory, United Kingdom

**Dellago, Christoph** - University of Vienna, Austria

**Farnudi, Ali** - Department of Physics, Sharif University of Technology, Iran

**Hammer, Valentin** - University of Vienna, Austria

**Kobsch, Anaïs** - CNRS, France

**Langini, Cassiano** - University of Zurich, Switzerland

**Li, Zhi** - University of Lyon (ENS), France

**Naleem, Nawavi** - New York University Abu Dhabi, United Arab Emirates

**O'Cais, Alan** - Jülich Supercomputing Centre, Germany

**Quemener, Emmanuel** - ENS de Lyon, France

**Reiner, Madlen** - University of Vienna, Austria

**Schaan, Renata** - ENS Lyon, France

**Singraber, Andreas** - University of Vienna, Austria

**Solomatova, Natalia** - ENS de Lyon, France

**Tavanti, Francesco** - University of Modena and Reggio Emilia, Italy

**Tribello, Gareth** - Queen's University Belfast, United Kingdom

# Electrochemical energy storage: Theory meets industry

Location: CECAM-FR-MOSER

Webpage: <https://www.cecarn.org/workshop-details/126>

Dates: Jun 12, 2019 - Jun 14, 2019

## 1 State of the art

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How much energy can a device store? How fast can it be charged? These two questions are at the heart of the research on electrochemical energy storage (EES). Two main families of devices coexist: supercapacitors which accumulate the charge at the surface of the electrodes through ion adsorption, and batteries in which the storage mechanism is based on redox reactions occurring in the bulk electrodes. Li-ion batteries have a high specific energy, keeping cellular phones, laptop and even cars working throughout several hours. For rapid power delivery and recharging, i.e. for high specific power applications, supercapacitors are then used. Due to the recent advances in the field of materials science, the range of applications of EES devices has tremendously increased over the past two decades. The development of systems with improved performances and lower costs, as well as their large-scale production are now considered as vital issues for many countries. This can be seen from the recent creation of networks or institutes that gather academics and industrials, both at the national and European levels. Most of the recent breakthroughs have however implied complex materials, often at the nanoscale. It is thus necessary to control the chemistry at the molecular level in all the active components of the devices, i.e. the two electrodes, the electrolytes. The various interfaces also have to be characterized and understood which implies considering potential dependent mechanistic approaches. Over the year, atomistic and molecular simulations have therefore appeared as one of the main keys to success in designing tomorrow's high-energy and high-power EES devices, in complement with in situ and/or in operando spectroscopy techniques. This is now well established in academic laboratories, which are now routinely building consortiums with synthesis, electrochemical and spectroscopic characterizations, together with modeling for developing new materials. However, this habit does not seem to be adopted yet by the industrial companies in the field. The objective of this workshop was therefore to bring together some of the worldwide experts in the field of EES simulations (and in particular the researchers who are developing the corresponding simulation tools) with the interested industrial partners. We hoped that such a workshop could help bridging the gap between needs and supply, which would put simulation at the centre of the future industrial developments of EES devices. The state-of-the-art can be considered at two levels: 1/ Simulation methods which are routinely used to simulate EES devices. 2/ Initiatives which are currently undertaken to bring simulation tools and/or results within the reach of non-specialist users. From the methodological point of view, many different methods are used or developed depending on the nature of the material, the targeted properties and the necessary time/length scales. -The workhorse for studying the redox activity of bulk electrode materials is standard Density Functional Theory (DFT) since it is necessary to have access to the electronic structure. -For electrolytes, determining the transport properties involves the use of molecular dynamics. Depending on the availability of correct force fields, classical or DFT-based MD are generally used. -Then further statistics or larger systems are generally studied by using lattice-based methods, such as kinetic Monte Carlo or Lattice Boltzmann. Generally, standard DFT or MD packages can be used to study bulk materials. However, in the case of interfaces, additional difficulties need to be overcome so that several groups are developing specific methodologies and/or simulation packages.

Despite the large growth in the simulation communities (especially DFT and MD) over the past decades, using these tools often requires lots of efforts for experimentalists and/or engineers in the industry. For this reason, several groups are currently developing user-friendly interfaces, either in specific programs or directly accessible from website. For efficiency reasons, it is necessary to develop high-throughput frameworks and to link these tools with accurate databases. This implies that a common language is established between the communities of theorists and experimentalists, in order to build appropriate databases that will be helpful for material designers. Finally, we should mention that several research groups are developing tools that aim to simulate systems at much larger scales. The objective is to provide a direct link with experiments, by directly computing macroscale properties similar to the ones obtained in electrochemistry experiments. Such multi-scale methods, most often based on the Butler-Volmer equation, are typically top-down approaches that aim to account for the material or electrolyte specificity in an effective manner through appropriate parameterizations. Huge efforts are being devoted to the development of bottom-up approaches, with however major issues due to transferability between different scales.

## 2 Major outcomes

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Two major scientific points were discussed in the meeting: 1/ Virtual screening The workshop was opened with an introductory lecture by Woomin Kyoung from Hyundai. He provided his company's view on how modeling should help the development of new materials. Although he discussed more specifically the case of electrolytes, he underlined that the process should be general for all the components of the battery. In short, the current approach is that new materials are discovered at the laboratory scale, then validated by using them in setups that are closer and closer to the commercial device. Overall, this discovery process takes up to 20 years. Hyundai believes that this time could be significantly reduced by involving modeling from the start, but it seems from the presentation that no procedure was clearly defined yet. This issue was then discussed in several presentations. In particular, G. Hautier (Univ. Catholique de Louvain) showed results from a computational screening procedure that led to the discovery of a new Li-ion battery material,  $\text{LiTi}_2(\text{PS}_4)_3$ , which shows promising performance. G. Hautier nevertheless pointed to the fact that there is still a need for a close interplay between simulations and experiments, the latter being very useful to control the screening procedure at various stages of developments. The difficulty when performing virtual screening is the choice of the descriptors to select the best materials, a problem which has been rather extensively tackled by groups working on electrocatalysis. Several presentations were dedicated to this topic, in particular A. Gross (Ulm University) showed the correlation between the height of self-diffusion barriers, derived from density functional theory calculations, and the occurrence of dendrite growth in batteries. T. Vegge (DTU energy) provided a number of recent examples of how DFT simulations supported by machine learning could be used efficiently to identify the limiting thermodynamic, ionic and electronic transport mechanisms in novel Li-ion electrode materials and Li-S batteries. 2/ Multi-scale modeling of Li-ion batteries Although the discovery of new materials is a major challenge, it is difficult to assess whether they can be commercialized based on a few intrinsic properties computed at the microscopic level. Indeed, most of the performance of the device depends on how the electrodes can be processed, put in shape, etc. These problems can be tackled by using a multi-scale modeling approach, whose principles were detailed in the lecture of A. Franco (Univ. of Picardie Jules Verne), who presented results from a collaboration with Renault. His approach combines discrete modeling techniques (kinetic Monte Carlo, Coarse Grained Molecular Dynamics, Lattice Boltzmann. . . ) with continuum models at multiple spatio-temporal scales and deep learning algorithms. He showed how this approach could be connected to experimental characterizations by focusing on the simulation of the manufacturing of lithium

ion batteries. A second illustration of the importance of multi-scale simulations was provided by C. Delacourt (Univ. of Picardie Jules Verne), who showed that it was possible to predict the occurrence of degradation phenomena, such as Li plating during charge, in order to better control (or avoid) it. In addition to these two points, all the presentations provided a good panorama of the simulation techniques available to simulate Li-ion batteries and supercapacitors, ranging from DFT to continuum methods. The feedback was very positive from the various industrial representatives, since some of them knew very little about modeling methods before the workshops.

## 3 Community needs

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The various presentations showed that there is a huge number of codes to simulate Li-ion batteries at all the scales. Nevertheless, several challenges still exist, in particular for the understanding of the electrode/electrolyte interface. Not surprisingly, most of the presentations underlined the need for large-scale HPC resources. In particular, virtual screening requires to perform simulations on thousands of materials, which cannot be made on local computers. It seems that the most important lack is currently the database. Even if there are several initiatives such as the Materials Genome, which was introduced by A. Van der Ven (University of California Santa Barbara), the various pre presentations showed how the simultaneous utilization of data from multiple domains (and not only simulation) plays a critical role in accelerating the industrial discovery process. More precisely, the accelerated discovery and inverse design of future batteries using generative deep learning models trained on all sources of available data, i.e., largescale multi-fidelity data sets, multi-scale computer simulations and databases, operando characterization from large-scale research facilities, high-throughput synthesis and laboratory testing will be needed very quickly. It would be interesting to discuss whether the CECAM could play a role on the development of such a database, since it is one of the only European organization which includes multi-scale modeling as an objective and not only a subpart of it (several centers of excellence were dedicated to materials but they were mostly interested in the use of DFT to study them). Anyway, such an initiative should strongly include the participation of experimentalists.

## 4 Funding

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During the meeting we mainly focused on the European project Battery 2030+, which has just been launched in 2019. This flagship program includes most of the main European universities and research centers, and it is supported by many organisations (see <https://battery2030.eu>). T. Vegge, who is implied in the proposal writing, has devoted a large part of his talk to present the objectives and how they should be tackled by the computational community. In particular, we should be implied in BIG-MAP: Battery Interface Genome - Materials Acceleration Platform. Several actions will be funded in the project, among which the most interesting for our community are: -The built of databases -The inverse computational design of battery materials and interface -Machine learning modules for automated analysis -Multiscale simulations and physical models All the results will be put together in a central Artificial Intelligence. It is clear that this project will be a very important opportunity of funding for European researchers working in the field of energy storage over the few next years.

## 5 Will these developments bring societal benefits?

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There are two main societal benefits of the development of better electrochemical energy storage devices: 1/ A better inclusion of renewable energies in the electricity mix. Indeed, due to their intermittent character, it is very difficult to fully benefit from the production of wind mills or solar panels in highly electrified regions such as Europe. They are very often complemented by fossil fuels-based production, which is highly problematic in terms of the carbon footprint. Nevertheless, these applications need the development of low-cost technologies in order to be competitive. Deploying these technologies is very urgent since global warming is already impacting our societies. 2/ Accelerating the electrification of the transport sector. The latter is responsible for a large share of the CO<sub>2</sub> emissions in European countries. The rapid growth of the Li-ion batteries has allowed electric vehicles to progressively increase their share on the market. In countries where the electricity is low-carbon (France, Norway, Belgium, Switzerland, etc), the impact on the carbon emissions should be very important on the future. These two benefits reflect on the list of industrial partners who participated to the workshop: Hyundai, Peugeot (PSA), Umicore, CEA, IFP, Happy electrons & Equinor.

## 6 Participant list

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### Organizers

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#### **Doublet, Marie-Liesse**

Institut Charles Gerhardt, France

#### **Salanne, Mathieu**

Sorbonne Université, France

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**Cabelguen, Pierre-Etienne** - Umicore, France

**Clement, Raphael** - University of California Santa Barbara, United States

**Delacourt, Charles** - Université de Picardie Jules Verne, France

**Franco, Alejandro A.** - Laboratoire de Reactivite et Chimie des Solides, Université de Picardie Jules Verne, CNRS UMR 7314, Amiens, France

**Ghadrdan, Maryam** - Systems Solutions, Norway

**Gosika, Mounika** - Indian Institute of Science, British Indian Ocean Territory

**Gross, Axel** - Ulm University, Germany

**Hautier, Geoffroy** - Université catholique de Louvain (UCL), Belgium

**Honkala, Karoliina** - University of Jyväskylä, Finland

**Kirchner, Barbara** - University of Bonn, Germany

**Kyoung, Woomin** - Hyundai Motor Company, Republic of Korea

**Martirez, Mark** - Princeton University, United States

**Merlet, Celine** - CNRS - Université Paul Sabatier, France

**Morgan, Ben** - Department of Materials, Oxford University, United Kingdom

**Van der Ven, Anton** - University of California Santa Barbara, United States

**Vegge, Tejs** - Department of Energy Conversion, Technical University of Denmark, Denmark

**Zhang, Chao** - Uppsala University, Sweden



# Extended software development workshop: quantum dynamics

Location: Durham University

Webpage: <https://www.cecarn.org/workshop-details/145>

Dates: Jul 08, 2019 - Jul 19, 2019

## 1 State of the art

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Quantum molecular dynamics simulations are pivotal to understanding and predicting the microscopic details of molecules, and strongly rely on a combined theoretical and computational effort. When considering molecular systems, the complexity of the underlying equations is such that approximations have to be devised, and the resulting theories need to be translated into algorithms and computer programs for numerical simulations. In the last decades, the joint effort of theoretical physicists and quantum chemists around the challenges of quantum dynamics made it possible to investigate the quantum dynamics of complex molecular systems, with applications ranging from energy conversion, energy storage, organic electronics, light-emitting devices, biofluorescent molecules, or photocatalysis, to name a few. Two different strategies have been successfully applied to perform quantum molecular dynamics: wavepacket propagation or trajectories. The first family of methods includes all quantum nuclear effects, but their computational cost hampers the simulation of systems with more than 10-12 degrees of freedom. The second family of methods introduces the idea of trajectories as a support to sample the nuclear configuration space, and can be divided, in turn, into two families: the so-called "trajectory basis functions" methods (as the variational multiconfigurational Gaussian or multiple spawning), and the quantum-classical methods (as Ehrenfest or surface hopping). A few weeks before the ESDW on Quantum Dynamics in Durham, an ECAM state-of-the-art workshop was held in Lyon (France). Some of the topics discussed in Lyon have been presented in more detail at the ESDW, in particular to highlight the relationship between the fundamental theory and the computational developments of the modules that will be presented by the end of 2019.

## 2 Training provided

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Different sessions with scientific presentations were organized to offer the opportunity to a large number of participants to discuss their work, in relation to the software developments proposed at the ESDW. With this idea in mind, we assigned 60-minute presentations to senior participants and postdoctoral researchers, and 30-minute presentations to PhD students. These scientific presentations focused largely on applications of wavepacket propagation methods and trajectory-based techniques, for which training was provided by experts in the fields (see below).

Many successful applications of such methods were highlighted, but at the same time some limitations and the problems encountered by the developers discussed. Both presenters and audience were composed of people strongly engaged in theory and numerical developments, therefore interesting discussions on various points arose during and after the presentations.

Among others:

- Performing calculations based on the trajectory surface hopping algorithm accounting explicitly for a laser field;

- Inclusion of spin-orbit interactions in the exact factorization formalism and the quantumclassical algorithm derived from it;
- Development of model potential energy surfaces (including spin-orbit coupling) for laserdriven dissociation of IBr;
- Limitations of the trajectory surface hopping algorithm, with and without decoherence corrections, in various molecular processes.

Concerning the training part of the ESDW, various aspects were covered

1. Theory: wavepacket propagation and trajectory-based methods;
2. Code development: ANVIL for continuous integration, GitLab, CUDA for GPU programming;
3. HPC: parallel computing with MPI.

On average, about 20 people attended the training sessions.

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## 3 List of the software development projects

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The modules discussed and developed during the ESDW are the following:

1. Quantics OpenMP (designed to perform HPC)
2. Exact factorization analysis
3. PaPIM QTB
4. CISTunfti
5. GuessSOC
6. Quantics DB OpenMP (designed to perform HPC)
7. PerGauss

These modules will be documented by the developers and finalized by November 2019 to be submitted as deliverables. For almost all modules, a merge request has been opened on the GitLab account of the ECAM developers.

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## 4 Future plans

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The follow-up meeting of the ESDW will be held at Durham University to discuss progress and prepare the following ESDW, most likely at the beginning of 2020. All modules only need to be finalized and are currently proposed by people who have been involved in the ECAM project already for a few years. Therefore, we believe that the deadline for the submission of the deliverables will be met without the need for an additional meeting.

## 5 Participant list N/A

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### Organizers

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**Agostini, Federica**

Université Paris-Sud, France

**Curchod, Basile**

Durham University, United Kingdom

**Worth, Graham**

University College London, United Kingdom

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# Challenges in Multiphase Flows

Location: Monash University Prato Center, Tuscany, see <http://monash.it/>

Webpage: <https://www.cecarn.org/workshop-details/152>

Dates: Dec 09, 2019 - Dec 12, 2019

## 1 State of the art

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The topic was mesoscale physics where at least partly a continuum (hydrodynamic) description is appropriate, and where interfaces play an important role. The strength of the meeting was its interdisciplinary approach, bringing together top experts from physics, engineering and mathematics. The school part provided a thorough overview over the existing concepts and methods, while the workshop discussed cutting-edge applications. Some highlights of the school, which provided interesting learning experiences even for us as experienced researchers, were the lucid explanation of the interface width as an adjustable parameter in diffuse-interface models, the systematic application of GENERIC to construct equations of motion, and the systematic upscaling of models for flow in porous media. Interesting developments at the workshop included, for example, the study of surface instabilities for viscoelastic fluids, systematic investigations of bubble nucleation via Fluctuating Hydrodynamics, and a clear outline of the heterogeneous multiscale method, which couples Molecular Dynamics with continuum mechanics.

## 2 Major outcomes

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We believe the most important outcome was the learning experience for everybody, and most prominently so for the students. Highlights have already been mentioned in the previous section.

## 3 Community needs

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We do feel that interdisciplinary networking is beneficial for the field, and our meeting has tried to address that. However, we do not think that the field is developing so rapidly that a series of workshops were appropriate. A clear-cut need of infrastructure in terms of HPC resources and/or codes was not identified. We feel that the conceptual problems in the field still prevail over the infrastructural ones, and routine push-the-button applications rarely exist.

## 4 Funding

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The meeting was funded by

- CECAM Headquarters (this was an ECAM state-of-the-art workshop within the mesoscale simulations work package),
- the Max Planck Institute for Polymer Research within its commitment for the SMSM node.

## 5 Will these developments bring societal benefits?

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This was about basic research. Nevertheless, there is clear industrial interest in this kind of research (for example, most obviously so in the studies of flow in porous media, which are important for oil recovery). However, apparently this does not go so far that industry would commit itself here to any measurable degree (and certainly not in terms of funding).

## 6 Participant list

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### Organizers

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#### **Duenweg, Burkhard**

Max Planck Institute for Polymer Research, Germany

#### **Jagadeeshan, Ravi Prakash**

Monash University, Melbourne, Australia

#### **Pagonabarraga, Ignacio**

CECAM, Switzerland

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**Eiser, Erika** - University of Cambridge, United Kingdom

**Ellero, Marco** - College of Engineering, University of Swansea, United Kingdom

**Harting, Jens** - Forschungszentrum Jülich, Germany

**Hassanizadeh, Majib** - Department of Earth Sciences, Utrecht University, Netherlands

**Hu, Xiangyu** - Technical University Munich, Germany

**Kreiss, Gunilla** - University Uppsala, Sweden

**Krueger, Timm** - University of Edinburgh, United Kingdom

**Kuipers, Hans** - Eindhoven University of Technology, Netherlands

**Lukacova, Maria Lukacova** - Institute of Mathematics, Germany

**Plapp, Mathis** - Ecole Polytechnique, Centre National de la Recherche Scientifique, France

**Schmid, Friederike** - Johannes Gutenberg University Mainz, Germany

**Yeomans, Julia** - Oxford University, United Kingdom



**CECAM 50  
CONFERENCE  
REPORT**



# Programme

DAY 1: MONDAY 9 SEPTEMBER 2019				
12:00-13:45	REGISTRATION			
13:45-14:35	<b>Room: LAUSANNE</b> Chairperson: Christoph Dellago <b>Hardy Gross (Plenary Speaker)</b> The Hebrew University of Jerusalem, Israel <b>Potential Energy Surfaces and Berry Phases from the Exact Factorization: A Predictive First-Principles Approach to Non-Adiabatic Dynamics</b>			
	<b>Room: ORSAY</b>	<b>Room: BLARICUM</b>	<b>Room: LAUSANNE</b>	<b>Room: LYON</b>
	<b>First Principle Materials Science and High Throughput</b> Chairperson: Leon Petit	<b>Statistical Physics and Physical Chemistry</b> Chairperson: Godehard Sutmann	<b>Algorithm Development and Mathematical Foundations</b> Chairperson: Carsten Hartmann	<b>Quantum and Ab-Initio Dynamics</b> Chairperson: David Coker
14:45-15:15	<b>Matthias Scheffler (Invited Speaker)</b> Fritz Haber Institute for the Structure and Dynamics of Matter, Berlin, Germany <b>Big Data and Artificial Intelligence in Materials Science: When The New Science Is in The Outliers</b>	<b>Angelos Michaelides (Invited Speaker)</b> University College London, United Kingdom <b>Interfacial Water: from Atmospheric Ice Nucleation to Nano-Confinement</b>	<b>Mark Tuckerman (Invited Speaker)</b> New York University, USA <b>Molecular Simulation and Machine Learning as Routes to Exploring Structure and Phase Behavior in Atomic and Molecular Crystals</b>	<b>Roi Baer (Invited Speaker)</b> The Hebrew University of Jerusalem, Israel <b>Unraveling Nonequilibrium Dynamics of Non-Interacting Electrons in Open Systems</b>
15:15-15:45	<b>Angel Rubio (Invited Speaker)</b> Max Planck Institute for the Structure and Dynamics of Matter, Mainz, Germany <b>Quantum-Cavity and Floquet-Engineered New States of Matter from a QEDFT Perspective</b>	<b>Lyderic Bocquet (Invited Speaker)</b> Ecole Normale Supérieure (ENS), Paris, France <b>Mechano-Sensitive Ion Conduction in Nanochannels</b>	<b>Benedict Leimkhuler (Invited Speaker)</b> University of Edinburgh, United Kingdom <b>Exploring Complex Landscapes: from Molecular Dynamics to Neural Networks</b>	<b>Aaron Kelly (Invited Speaker)</b> Dalhousie University, Canada <b>Nonequilibrium Charge and Energy Transport</b>
15:45-16:05	<b>Ralph Gebauer (Contributed Talk)</b> International Centre for Theoretical Physics, Trieste, Italy <b>Water Splitting on Hematite Surfaces: Insights from Density-Functional Theory</b>	<b>Alberto Giacomello (Contributed Talk)</b> University of Rome 'La Sapienza', Italy <b>Perpetual Superhydrophobicity: Drying from the Nanoscale to the Macroscale</b>	<b>Pietro Faccioli (Contributed Talk)</b> University of Trento, Italy <b>All-Atom Simulation of Protein Folding and Direct Validation Against Time-Resolved Spectroscopy Experiments</b>	<b>Lidice Cruz Rodriguez (Contributed Talk)</b> Research Institute on Complex Atomic and Molecular Systems, Toulouse, France <b>Trajectory-Based Method for the Study of Ultrafast Quantum Dynamics.</b>
16:05-16:35	COFFEE BREAK			

	<b>Room: ORSAY</b>	<b>Room: BLARICUM</b>	<b>Room: LAUSANNE</b>	<b>Room: LYON</b>
	<b>High-Throughput and Big Data</b>	<b>Statistical Physics and Physical Chemistry</b>	<b>Algorithm Development and Mathematical Foundations</b>	<b>Soft Matter</b>
16:35-16:55	<b>Jutta Rogal (Contributed Talk)</b> Ruhr-University Bochum, Germany <b>Neural Network-Based Path Collective Variables for Enhanced Sampling of Phase Transformations</b>	<b>Felix Höfling (Contributed Talk)</b> Free University of Berlin, Germany <b>Emergence of Molecular Friction in Liquids: A Memory-Kernel Perspective</b>	<b>Erik E. Santiso (Contributed Talk)</b> North Carolina State University, USA <b>Modeling Crystal Nucleation with the String Method</b>	<b>Gerhard Kahl (Contributed Talk)</b> Vienna University of Technology, Austria <b>Dendrimer-Like DNAs: Simulation and Experiment</b>
16:55-17:15	<b>Daniele Padula (Contributed Talk)</b> Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland <b>Singlet Fission Molecules among Known Compounds: Finding a Few Needles in a Haystack</b>	<b>Céline Merlet (Contributed Talk)</b> CNRS-University of Toulouse, France <b>Simulation of NMR Spectra for Molecules or Ions Diffusing in Porous Disordered Carbons</b>	<b>J. Zhang (Contributed Talk)</b> Peking University, Beijing, China <b>Learning Clustered Representation for Complex Free Energy Landscapes</b>	<b>Jure Dobnikar (Contributed Talk)</b> Chinese Academy of Sciences, Beijing, China University of Cambridge, United Kingdom <b>Emerging Interactions through Active Alignment</b>
17:15-17:35	<b>Max Veit (Contributed Talk)</b> Swiss Federal Institute of Technology (EPFL), Lausanne, Switzerland <b>Machine Learning Potentials for Molecular Liquids</b>	<b>Matteo Ceccarelli (Contributed Talk)</b> University of Cagliari, Italy <b>Multiscale Modeling of Molecular Diffusive Transport Phenomena: from Microscopic Details to Macroscopic Fluxes</b>	<b>Mauricio J. Del Razo (Contributed Talk)</b> Free University of Berlin, Germany <b>Modeling and Simulating Molecular Kinetics as Diffusion Processes with Markovian Switching</b>	<b>Silvia Corezzi (Contributed Talk)</b> University of Perugia, Italy <b>Autocatalytic Patchy Particles</b>
17:40-18:30	<b>Room: LAUSANNE</b> Chairperson: Michel Mareschal <b>Julia Yeomans (Plenary Speaker)</b> University of Oxford, United Kingdom <b>Topology in Biology?</b>			



DAY 2: TUESDAY 10 SEPTEMBER 2019				
9:00-9:50	Room: LAUSANNE		Chairperson: Giovanni Ciccolti	
	Giulia Galli ( <i>Plenary Speaker</i> ) University of Chicago, USA			
9:50-10:30	COFFEE BREAK			
	Room: ORSAY	Room: BLARICUM	Room: LAUSANNE	Room: LYON
	Electronic Structure – First Principle Modelling Chairperson: Paul Durham	Soft Matter Chairperson: Kurt Kremer	Biophysics and Biochemistry Chairperson: Francesco Gervasio	Statistical Physics and Physical Chemistry Chairperson: Rodolphe Vuilleumier
10:30-11:00	Lucia Reining ( <i>Invited Speaker</i> ) Ecole Polytechnique, Palaiseau, France  Calculation of Electronic Spectra Using Auxiliary Systems and Model Results	Marcus Müller ( <i>Invited Speaker</i> ) Georg-August University, Göttingen, Germany  Structure Formation in Copolymer Materials After Sudden Quenches	Erik Lindahl ( <i>Invited Speaker</i> ) Stockholm University, Sweden  Simulations of Structural Transformations in Ceramide Phases from Fitting Structures to CEMOVIS Image Data	Benjamin Rotenberg ( <i>Invited Speaker</i> ) Sorbonne University, Paris, France  "Use the Force!" Reduced Variance Estimators for Radial Distribution Functions, Generic 3d Densities and (Local) Transport Coefficients
11:00-11:30	Kristian Sommer Thygesen ( <i>Invited Speaker</i> ) Technical University of Denmark, Kongens Lyngby Denmark  Data-Driven Discovery of Novel Two-Dimensional Materials	Markus Deserno ( <i>Invited Speaker</i> ) Carnegie Mellon University, USA  The Role of Dynam Twist in Membrane Fission	Ron Elber ( <i>Invited Speaker</i> ) The University of Texas, Austin, USA  Computer Simulations of Biological Systems: Proteins, RNA and Membranes	Kirsten Martens ( <i>Invited Speaker</i> ) University Grenoble Alpes, France  Modeling Approaches for Soft Glassy Rheology
11:30-12:00	Kersti Hermansson ( <i>Contributed Talk</i> ) Uppsala University, Sweden  OHI	Ludovic Berthier ( <i>Invited Speaker</i> ) University of Montpellier, France  Equilibrium Simulations of Supercooled Liquids Beyond Laboratory Timescales	Davide Marenduzzo ( <i>Invited Speaker</i> ) University of Edinburgh, United Kingdom  Biophysical Principles of Transcription-Driven Chromosome Organisation	Nuno Araujo ( <i>Invited Speaker</i> ) University of Lisbon, Portugal  Self-Folding Kirigami at the Microscale
12:00-12:20	Enrico Tapavicza ( <i>Contributed Talk</i> ) California State University Long Beach, USA  First-Principles Prediction of Vibronic Spectra, Internal Conversion and Wavelength-Dependent Product Quantum Yields	Marisol Ripoll ( <i>Contributed Talk</i> ) Jülich Research Centre, Germany  Phoretic Colloids and Micromachines	Marie-Christine Sawley ( <i>Contributed Talk</i> ) Intel Semiconductor Ag, Zurich, Switzerland  25 Years of Biosimulation	Aleksandar Donev ( <i>Contributed Talk</i> ) Courant Institute, New York University, USA  Fluctuating Hydrodynamics of Electrolytes
12:20-12:40	M. V. Ganduglia-Pirovano ( <i>Contributed Talk</i> ) Institute of Catalysis and Petrochemistry-CSIC, Madrid, Spain  Oxygen-Vacancy Ordering and Dynamics at the Reduced CeO <sub>2</sub> (111) Surface and the Entanglement with Polaron Hopping	Angelo Rosa ( <i>Contributed Talk</i> ) International School of Advanced Studies – SISSA, Trieste, Italy  From Chromosome Territories to Ring Polymers: Physical Properties of Untangled Polymers Melts	Alberto Pérez de Alba Ortiz ( <i>Contributed Talk</i> ) University of Amsterdam, The Netherlands  Simultaneous Sampling of Multiple Transition Channels Using Adaptive Paths of Collective Variables	Daniel Borgis ( <i>Contributed Talk</i> ) CNRS-Ecole Normale Supérieure De Paris (ENS), France Maison De La Simulation, Saclay, France  Ion-Induced Long-Range Orientational Correlations in Highly Diluted Electrolytes: What Do Second Harmonic Scattering Experiments Measure?

LUNCH BREAK				
12:45-14:15	LUNCH BREAK			
14:15-15:05	Room: LAUSANNE		Chairperson: Berend Smit	
	Werner Krauth ( <i>Plenary Speaker</i> ) Ecole Normale Supérieure (ENS), Paris, France  Fast Irreversible Markov Chains in Statistical Physics			
	Room: ORSAY	Room: BLARICUM	Room: LAUSANNE	Room: LYON
	Materials Modelling Chairperson: Magali Benoit	Soft Matter Chairperson: Jean Louis Barrat	Statistical Physics and Physical Chemistry Chairperson: Amir Natan	Biophysics and Biochemistry Chairperson: Majdi Hochlaf
15:05-15:35	Mariana Rossi ( <i>Invited Speaker</i> ) Fritz Haber Institute of the Max Planck Society, Berlin, Germany  In silico Synthesis of Microgels: Structure, Elasticity and Effective Interactions in Bulk and at Liquid-Liquid Interfaces	Emanuela Zaccarelli ( <i>Invited Speaker</i> ) CNR- Institute for Complex Systems, Rome, Italy  The Physics of Smarter and More Sustainable Cements	Emanuela Del Gado ( <i>Invited Speaker</i> ) Georgetown University, Washington DC, USA  The Physics of Crumpling and Folding of Untangled Polymers and Chromosomes	Ralf Everaers ( <i>Invited Speaker</i> ) Ecole Normale Supérieure (ENS), Lyon, France  Emergence of Knots in Intrinsically Disordered Proteins
15:35-16:05	Hannes Jonsson ( <i>Invited Speaker</i> ) University of Iceland, Reykjavik  Rate Theory for Magnetic Transitions with Application to Skyrmions and Molecular Magnets	Marjolein Dijkstra ( <i>Invited Speaker</i> ) Debye Institute, University of Utrecht, Netherlands  Birth of a Binary Crystal Nucleus of Hard Spheres	Gerhard Gompper ( <i>Invited Speaker</i> ) Jülich Research Centre, Germany  Active Filaments, Membranes, and Cells	Marek Cieplak ( <i>Invited Speaker</i> ) Polish Academy of Science, Warsaw, Poland  Prebiotic Chemistry and Origins of Life Studies Through Ab Initio Calculations
16:05-16:35	COFFEE BREAK			
16:35-16:55	Stefano Mossa ( <i>Contributed Talk</i> ) Interdisciplinary Research Institute of Grenoble (IRIG), France  Vibrational Excitations and Elastic Response at the Nanoscale: from Disordered Solids to Nanostructures	Francisco Vega Reyes ( <i>Contributed Talk</i> ) University of Extremadura, Spain  Complex Memory in Soft Materials	William A. Curtin ( <i>Contributed Talk</i> ) Swiss Federal Institute of Technology (EPFL), Lausanne, Switzerland  From Density Functional Theory to Strength and Ductility of Complex Metal Alloys	A. Marco Saitta ( <i>Contributed Talk</i> ) Sorbonne University, Paris, France  Challenges, Advances and Perspectives of Protein Aggregation Simulations
16:55-17:15	Marcella Iannuzzi ( <i>Contributed Talk</i> ) University of Zurich, Switzerland  Molecules at the Electrochemical Interface: Understanding Experiments with Simulations	Jean Paul Ryckaert ( <i>Contributed Talk</i> ) University of Brussels, Belgium  Shape, Height Fluctuations and Resisting Force of a Membrane Deformed by an Actin Bundle	Kostas Daoulas ( <i>Contributed Talk</i> ) Max Planck Institute for Polymer Research, Mainz, Germany  Symmetry-Inspired Mesoscopic Models for Studying Partially-Ordered Mesophases of Semiconducting Polymers	Birgit Strodel ( <i>Contributed Talk</i> ) Jülich Research Centre, Germany  Challenges, Advances and Perspectives of Protein Aggregation Simulations
17:25-18:15	Room: LAUSANNE		Chairperson: Wanda Andreoni	
	Gabor Csanyi ( <i>Plenary Speaker</i> ) University of Cambridge, United Kingdom  Advances in Interatomic Potentials for Materials			

DAY 3: WEDNESDAY 11 SEPTEMBER 2019				
9:00-9:50	Room: LAUSANNE		Chairperson: Dominic Tidesley	
	Ursula Röthlisberger ( <i>Plenary Speaker</i> ) Swiss Federal Institute of Technology (EPFL), Lausanne, Switzerland			
9:50-10:30	COFFEE BREAK			
	Room: ORSAY	Room: BLARICUM	Room: LAUSANNE	Room: LYON
	Electronic Structure – First Principle Modelling Chairperson: Thierry Deutsch	Statistical Physics Chairperson: Burkhard Duenweg	Biophysics and Biochemistry Chairperson: Vittorio Limongelli	Physical Chemistry Chairperson: Florent Calvo
10:30-11:00	Silvana Botti ( <i>Invited Speaker</i> ) Friedrich Schiller University, Jena, Germany  Exchange-Correlation Functionals for the Band Structure of Solids: from Bulk Systems to Interfaces and Surfaces	Chantal Valeriani ( <i>Invited Speaker</i> ) Complutense University of Madrid, Spain  The Role Played by Interactions in the Assembly of Active Colloids	Ilpo Vattulainen ( <i>Invited Speaker</i> ) Tampere University of Technology, Finland  Biosimulations in Life Sciences: Coarse-Grained Molecular Simulation Models Are Usually Superb, But When Do We Dare to Use Them?	M. Sprik ( <i>Invited Speaker</i> ) University of Cambridge, United Kingdom  Electromechanics of the Liquid Water-Vapour Interface
11:00-11:30	Feliciano Giustino ( <i>Invited Speaker</i> ) University of Oxford, United Kingdom  Looking Inside a Polaron	Matthieu Wyart ( <i>Invited Speaker</i> ) Swiss Federal Institute of Technology (EPFL), Lausanne, Switzerland  A Jamming Transition Affects Landscape and Generalisation in Deep Learning	Modesto Orozco ( <i>Invited Speaker</i> ) University of Barcelona, Spain  DNA a 10 <sup>10</sup> Multiscale Problem	Marialore Sulpizi ( <i>Invited Speaker</i> ) Johannes Gutenberg University Mainz, Germany
11:30-12:00	Georg Kresse ( <i>Invited Speaker</i> ) University of Vienna, Austria  Hybrid Perovskites: From the Random Phase Approximation to Machine Learned Potentials	Srikanth Sastri ( <i>Invited Speaker</i> ) Jawaharlar Nehru Centre for Advanced Scientific Research (JNCASR), Bangalore, India  Self Organization and Irreversibility in Cyclically Driven Amorphous Matter	Peter Bolhuis ( <i>Invited Speaker</i> ) University of Amsterdam, Netherlands  Understanding the Dynamical Bottlenecks in Complex Activated (Bio)Molecular Processes	Mathieu Salanne ( <i>Invited Speaker</i> ) Sorbonne University, France  Electrode Models for Computational Electrochemistry
12:00-12:20	Göran Wendin ( <i>Contributed Talk</i> ) Chalmers University of Technology, Sweden  Calculating Ground State Energy Surfaces for H <sub>2</sub> O and HCN with a Quantum Computer	Matthias Fuchs ( <i>Contributed Talk</i> ) University of Konstanz, Germany  Emergence of Long-Ranged Stress and Strain Correlations in Viscoelastic Liquids	Elise Dumont ( <i>Contributed Talk</i> ) Ecole Normale Supérieure (ENS), Lyon, France  Mapping DNA-Photosensitizers Interaction in and Out the Nucleosome	Carlo Pierleoni ( <i>Contributed Talk</i> ) University of L'Aquila, Italy  High Pressure Liquid Hydrogen Across Molecular Dissociation
12:20-12:40	Zhao Wang ( <i>Contributed Talk</i> ) Tu Wien, Vienna, Austria Guangxi University, China  Selective Conduction of Organic Molecules on Free-Standing Graphene and Carbon Nanotubes	Giuseppe Foffi ( <i>Contributed Talk</i> ) University of South Paris, France  Slowing Down Supercooled Liquids by Manipulating their Local Structure	Anton Polyansky ( <i>Contributed Talk</i> ) University of Vienna, Austria  Towards an Atomistic Structure of Phase-Separated Protein Granules	Saikat Datta ( <i>Contributed Talk</i> ) University of Edinburgh, United Kingdom  Nanobubble Nucleation Due to Surface Vibration
12:45-14:15	LUNCH BREAK			

CECAM 50 CELEBRATION	
	Room: LAUSANNE Chairperson: Alessandro Laio
14:00-14:30	Daan Frenkel University of Cambridge, United Kingdom  Lecture on CECAM History, Role and Contributions
14:30-15:45	CECAM Berni Alder prize ceremony
15:45-16:15	COFFEE BREAK
	Room: LAUSANNE Chairperson: Ignacio Pagonabarraga
16:15-18:00	Round table  Silke Biermann Ecole Polytechnique, Palaiseau, France Nicola Marzari Swiss Federal Institute of Technology (EPFL), Lausanne, Switzerland Elisa Molinari University of Modena and Reggio Emilia, Italy Michele Parrinello ETH Zürich and USI Lugano, Switzerland Christof Schütte Zuse Institute Berlin, Germany Matthias Troyer One Microsoft Way Redmond, USA Martin Vetterli Swiss Federal Institute of Technology (EPFL), Lausanne, Switzerland
19:00-00:00	CONFERENCE DINNER AT THE OLYMPIC MUSEUM

DAY 4: THURSDAY 12 SEPTEMBER 2019				
9:00-9:50	Room: LAUSANNE			Chairperson: Stefano Baroni
	<b>Ali Alavi (Plenary Speaker)</b> Max Planck Institute for Solid State Research, Stuttgart, Germany <b>Non-Unitary and Stochastic Quantum Chemistry</b>			
9:50-10:30	COFFEE BREAK			
	Room: ORSAY	Room: BLARICUM	Room: LAUSANNE	Room: LYON
	<b>Electronic Structure and Materials Modelling</b> Chairperson: Fabio Affinito	<b>Statistical Physics and Physical Chemistry</b> Chairperson: Filippo de Angelis	<b>Biophysics and Biochemistry</b> Chairperson: Donal Mackernan	<b>Physical Chemistry</b> Chairperson: Matej Praprotnik
10:30-11:00	<b>Roman Martonak (Invited Speaker)</b> Comenius University, Bratislava, Slovakia  <b>Quantum and Classical Ripples in Graphene</b>	<b>Doros Theodorou (Invited Speaker)</b> National Technical University, Athens, Greece  <b>Atomistic and Mesoscopic Simulations of Polymer Melts</b>	<b>Simone Meloni (Invited Speaker)</b> University of Ferrara, Italy  <b>Multiscale Simulations of Nanofluidic Porous Systems for Energy Applications</b>	<b>Michele Carloti (Invited Speaker)</b> Swiss Federal Institute of Technology (EPFL), Lausanne, Switzerland  <b>Machine Learning for Atomic and Molecular Simulations</b>
11:00-11:30	<b>Maria Celia Righi (Contributed Talk)</b> University of Modena and Reggio Emilia, Modena, Italy  <b>Monitoring Water Splitting at Graphene Edges: Insights Into the Effects of Humidity on the Lubricity of Graphitic Materials</b>	<b>Francesco Mauri (Invited Speaker)</b> University of Rome La Sapienza, Italy  <b>Unified Theory of Thermal Transport in Crystalline Solids and Glasses</b>	<b>Ryoichi Yamamoto (Invited Speaker)</b> University of Kyoto, Japan  <b>Impact of Wall Constraint on the Dynamics of Self-Propelled Particles</b>	<b>Tanja Schilling (Invited Speaker)</b> University of Freiburg, Germany  <b>Coarse-Graining Out of Equilibrium</b>
11:30-11:50	<b>Mauro Causa (Contributed Talk)</b> University of Naples Federico II, Italy  <b>The Maximum Probability Domains (MPD) As a Powerful Tools for Analysing Electronic Structures</b>	<b>Stephen J. Cox (Contributed Talk)</b> University of Cambridge, United Kingdom  <b>Stabilization of AgI's Polar Surfaces by the Aqueous Environment, and its Implications for Ice Formation</b>	<b>Barak Hirshberg (Contributed Talk)</b> Swiss Federal Institute of Technology (ETHZ), Zurich, Switzerland  <b>Path Integral Molecular Dynamics for Cold Bosons</b>	<b>Grisell Diaz Leines (Contributed Talk)</b> Ruhr University, Bochum, Germany  <b>Maximum Likelihood Analysis of Reaction Coordinates during Crystal Nucleation in N</b>
11:50-12:10	<b>Tatsuhiko Ohto (Contributed Talk)</b> Osaka University, Japan  <b>Accessing the Accuracy of Density Functional Theory through Structure and Dynamics of the Water-Air Interface</b>	<b>Guido Roma (Contributed Talk)</b> University of Paris-Saclay, France  <b>Oxidation Products of Polyethylene and their Optical Signatures</b>	<b>Dennis R. Salahub (Contributed Talk)</b> University of Calgary, Canada  <b>Towards Free-Energy Profiles for Nano-Catalyzed Chemical Reactions in Complex Environments</b>	<b>Robinson Cortes Huerto (Contributed Talk)</b> Max Planck Institute for Polymer Research, Mainz, Germany  <b>Open Boundaries in Hamiltonian Adaptive Resolution Simulations: from Grand Canonical to Nonequilibrium Molecular Dynamics</b>
12:25-13:15	Room: LAUSANNE			Chairperson: Daan Frenkel
	<b>Eric Vanden-Eijnden (Plenary Speaker)</b> Courant Institute of Mathematical Sciences, New York University, USA <b>Molecular Dynamics Simulations in the Age of Machine Learning</b>			
13:20-13:30	CLOSING CEREMONY			



**List  
Of  
Abstracts**

## Plenary speakers

### ***Potential energy surfaces and berry phases from the exact factorization: A predictive first-principles approach to non-adiabatic dynamics***

**Eberhard Gross**

Max Planck Institute of Microstructure Physics, Halle - Germany

Room Lausanne, 13:45

Monday 9

Some of the most fascinating phenomena in physics and chemistry, such as the process of vision, exciton dynamics in photovoltaic systems, as well as phonon-driven superconductivity occur in the so-called non-adiabatic regime where the coupled motion of electrons and nuclei beyond the Born-Oppenheimer approximation is essential. The Born-Oppenheimer approximation is among the most fundamental ingredients of condensed-matter theory. It not only makes computations feasible, it also provides us with an intuitive picture of chemical reactions. Yet it is an approximation. To go beyond it is notoriously difficult because one has to start from the full many-body Hamiltonian of interacting electrons and nuclei. We deduce an exact factorization [1] of the full electron-nuclear wave function into a purely nuclear part and a many-electron wave function which parametrically depends on the nuclear configuration and which has the meaning of a conditional probability amplitude. The equations of motion for these two wave functions provide an ideal starting point to develop efficient algorithms for the study non-adiabatic phenomena. The successful prediction of laser-induced isomerisation processes [2], the description of decoherence [3], calculations of the molecular Berry phase without invoking the Born-Oppenheimer approximation [4] and accurate predictions of vibrational dichroism will demonstrate the power of this new approach. To tackle non-adiabatic phenomena in solids, such as laser-induced phase transitions, the equations of motion of the exact factorization are “density-functionalized” [5], leading to a coupled set of Kohn-Sham equations for electrons and phonons [6].

[1] A. Abedi, N.T. Maitra, E.K.U. Gross, *PRL* **105**, 123002 (2010).

[2] F. Agostini, S.K. Min, I. Tavernelli, E.K.U. Gross, *J Phys Chem Lett* **8**, 3048 (2017).

[3] S.K. Min, F. Agostini, E.K.U. Gross, *PRL* **115**, 073001 (2015).

[4] S.K. Min, A. Abedi, K.S. Kim, E.K.U. Gross, *PRL* **113**, 263004 (2014).

[5] R. Requist, E.K.U. Gross, *PRL* **117**, 193001 (2016).

[6] R. Requist, C.R. Proetto, E.K.U. Gross, *Phys Rev B* **99**, 165136 (2019).

### ***Topology in biology?***

**Julia Yeomans**

Oxford University, Oxford - United Kingdom

Room Lausanne, 17:40

Monday 9

Active materials, such as bacteria, molecular motors and self-propelled colloids continuously transform chemical energy from their environment to mechanical work. Dense active matter shows mesoscale turbulence, the emergence of chaotic flow structures characterised by high vorticity and self-propelled topological defects I shall describe how the ideas of active matter are suggesting new ways of interpreting cell motility and cell division. In particular recent results indicate that active topological defects may help to regulate turnover in epithelial cell layers and contribute to controlling the structure of bacterial colonies.

## Invited speakers

### ***Unraveling nonequilibrium dynamics of non-interacting electrons in open system***

**Roi Baer**

The Hebrew University of Jerusalem, Israel

**Room Lyon, 14:45**

**Monday 9**

The Lindblad equation describes quantum dynamics in systems that cannot be completely isolated from an environment, addressing dissipation and decoherence phenomena. Numerous applications of the formalism exist for small Hilbert-space systems, such as an atom or a molecule. But for many-electron systems, the dynamics are intractable even if Coulomb repulsion could be switched off since the particles would still be able to affect each other by interacting with the bath. Here, we develop an approximate approach for evolving non-interacting Fermions in open quantum systems based on the following elements:

- 1) The form of frequency-dependent Lindblad operators as formulated by Davies.
- 2) The unraveling procedure for converting the Lindblad equation into a stochastic Schrodinger equation.
- 3) Construction of time-dependent stochastic Lindblad operators.
- 4) Deploying the Hubbard-Stratonovich transformations [4-5] to represent the two-body interactions.
- 5) A collapsing procedure between time steps.

The method aims for open quantum systems that eventually include thousands of electrons and dense bands. Achieving this goal is wanting.

[1] Ruan, Z. and Baer, R. *Unravelling open-system quantum dynamics of non-interacting Fermions. Mol. Phys.* **116**, 2490-2496 (2018).

[2] Davies, E. B., *Markovian master equations, Communications in Mathematical Physics*, **39**, 91-110 (1974).

[3] Gisin, N., and Percival, I. C. *The Quantum-State Diffusion-Model Applied to Open Systems, Journal of Physics a-Mathematical and General*, **25**, 5677-5691, (1992).

[4] Hubbard J. *Calculation of Partition Functions, Phys. Rev. Lett.*, **3**, 77 (1959).

[5] Stratonovich, R. L. *A method for the computation of quantum distribution functions, Dokl. Akad. Nauk SSSR*, **115**, 1097 (1957).

### ***Mechano-sensitive ion nanochannels***

**Lyderic Bocquet**

Ecole Normale Supérieure, Paris - France

**Room Blaricum, 15:15**

**Monday 9**

Ion transporters in Nature exhibit a wealth of complex transport properties such as voltage gating, activation, or mechano-sensitive behavior. When combined, such processes result in advanced ionic machines achieving active ion transport, high selectivity, or signal processing. On the artificial side, there has been recent progress in the design and study of transport in ionic channels at nanoscales, but mimicking the advanced functionalities of ion transporters remains as yet out of reach. A prerequisite is the development of ionic responses sensitive to external stimuli.

In this talk, I will highlight various examples of strongly non-linear ionic transport in 1D and 2D nanofluidic channels, leading to mechano-sensitive ion conduction. I will first discuss experiments on ionic transport across 2D channels with sub-nanometer dimensions, made of van der Waals heterostructures. Experiments demonstrate a non-linear coupling between water flow and ionic transport, taking the form of a voltage gated streaming mobility.

Conversely I will show that (1D) carbon nanotubes with sub-2 nanometer size also highlight strongly non-linear response: however it rather takes the form of a pressure-dependent conductance, which mimicks very closely the response of biological mechano-sensitive ion channels.

Beyond I will explore how the specificities of transport at these scales allow for Coulomb blockade in ionic transport, demonstrating single ion transport and quantized ionic conduction at room temperature. Theory predicts a classical ionic quantization which results from the many-body interaction of ions in strong confinement coupled to a fractional Wien effect.

Altogether the specificity of nanoscale transport offers new leads to finely tune ion transport by mechanical stimuli. It provides a promising avenue for the design of tailored membrane functionalities.

*"Ionic Coulomb blockade as a fractional Wien effect"* N. Kavokine, S. Marbach, A. Siria, L. Bocquet, *Nature Nano* **14**, 573–578 (2019).

*"Molecular streaming and voltage-gated response in Angström scale channels"* T. Mouterde, A. Keerthi, A. Poggioli, S. Dar, A. Siria, A.K. Geim, L. Bocquet and R. Boya, *Nature* **567**, 87 (2019).

*"Dramatic pressure-sensitive ion conduction in conical nanopores"* L. Jubin, A. Poggioli, A. Siria and L. Bocquet, *Proc. Nat. Acad. Sci USA* **115** 4063-4068 (2018).

### **Self-interaction corrected functional calculations of molecules and solids**

**Hannes Jonsson**<sup>1,2</sup>

<sup>1</sup>University of Iceland, Reykjavik, Iceland

<sup>2</sup>Brown University, USA

**Room Orsay, 15:15**

**Monday 9**

While Kohn-Sham density functional theory (DFT) using GGA, meta-GGA and hybrid functional approximations has been remarkably successful in a variety of applications, there are several important cases where it fails. A variational and self-consistent implementation of the Perdew-Zunger self-interaction correction (PZ-SIC) using complex optimal orbitals has been applied to several such systems and found to give good results. Calculations of Mn dimer, dipole bound anion, Rydberg excited states of molecules and molecular clusters, localized charge state in a diamine cation and electronic holes in oxide crystals will be presented. The computational effort of the PZ-SIC calculations scales with system size in the same way as DFT/GGA calculations but the prefactor is large since an effective potential needs to be evaluated for each orbital (calculations that could, however, be carried out in parallel) and optimal orbitals need to be found in terms of the Kohn-Sham orbitals in each iteration. PZ-SIC is an example of an extended functional form where the energy depends explicitly on the orbital densities, not just the total electron density. While significant improvements are obtained with PZ-SIC compared with practical implementations of Kohn-Sham DFT, problems can also be introduced such as incorrect symmetry breaking. The orbital density dependent functional form could, however, be exploited more generally to develop a self-interaction free functional rather than as a correction to Kohn-Sham functionals, thereby providing a mean field theory for optimal orbitals and orbital energies.

### **Nonequilibrium charge and energy transport**

**Aaron Kelly**

Dalhousie University, Halifax - Canada

**Room Lyon, 15:15**

**Monday 9**

Recently developed approaches for simulating nonequilibrium quantum dynamics in nanoscale systems will be discussed. The unifying feature that these methods share is an ensemble of trajectories that is employed in order to construct observables and transport properties. We will explore the performance of selected techniques of this type in a variety of real-time quantum dynamics simulations of nonadiabatic charge and energy transfer processes, including cavity-bound spontaneous emission, charge separation and polaron formation donor-acceptor interfaces, and heat transport through molecular junctions.

### **Exploring complex landscapes: from molecular dynamics to neural networks**

**Benedict Leimkuhler**

University of Edinburgh, Edinburgh - United Kingdom

**Room Lausanne, 15:15**

**Monday 9**

Molecular dynamics (MD) is a versatile family of algorithms, largely designed for exploring statistical distributions (thermodynamic ensembles) in high dimensions. I will describe some recent success we have had in constructing Langevin-based stochastic numerical methods for accurate and stable sampling in various settings: for canonical sampling, for constant temperature and pressure, for constraints. But

there is no reason to restrict the application of these tools--they are extremely general, easy to implement, and ergodic (thus robust), and they can serve as the foundation for a rich collection of enhanced methods, e.g. tempering procedures and ensemble preconditioners. As discussed in other lectures at CECAM50, molecular dynamics is, increasingly, being combined with machine learning (e.g. neural networks) to improve flexibility and scalability. However, as I will show in this talk, machine learners can also gain insight from molecular dynamics. MD algorithms provide powerful tools for mapping the "loss landscape" of neural networks. In some cases MD algorithms can accelerate training procedures and provide information regarding the uncertainties of learned models. I will discuss our TATi system (Thermodynamic Analytics Toolkit) and its potential for use in data analytics.

### **Interfacial water: from atmospheric ice nucleation to nano-confinement**

**Angelos Michaelides**

University College London, London - United Kingdom

**Room Blaricum 14:45**

**Monday 9**

There are few molecules, if any, more important than water. Yet, major gaps in our molecular-level understanding of water persist, particularly for water at interfaces. In this talk I will discuss some of our recent work on water and ice. This will include new insights into the mechanisms of homogenous [1] and heterogenous ice nucleation [2-5] and the structure and dynamics of water at solid surfaces [6] and when confined within 2-dimensional membranes [7]. Methodological developments aimed at providing more accurate treatments of adsorption on and bonding within solids will also be covered [8-9], as well as a brief perspective on the future of the field.

[1] Fitzner et al., *Proc. Nat. Acad. Sci.* **116**, 2009 (2019).

[2] Kiselev et al., *Science* **355**, 367 (2017).

[3] Liriano et al., *J. Am. Chem. Soc.* **139** 6403 (2017).

[4] Fitzner et al., *Nature Comm.* **8**, 2257 (2017).

[5] Gerrard et al., *J. Am. Chem. Soc.* **141**, 8599 (2019).

[6] Hussain et al. *Nature Mater.* **16**, 461 (2016).

[7] Chen et al., *Phys. Rev. Lett.* **116**, 025501 (2016).

[8] Al-Hamdani et al., *J. Chem. Phys.* **147**, 044710 (2017).

[9] Zen et al., *Proc. Nat. Acad. Sci.* **115**, 1724 (2018).

### **Big data and artificial intelligence in materials science: when the new science is in the outliers**

**Matthias Scheffler, Luca M. Ghiringhelli**

Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin - Germany

**Room Orsay, 14:45**

**Monday 9**

Several issues hamper progress in data-driven computational science. In particular, these are a missing **FAIR** [1] **data infrastructure** and appropriate **data-analytics methodology** [2].

Significant efforts are still necessary to fully realize the A and I of FAIR. Here the development of metadata, their intricate relationships, and data ontology need more attention. In this talk, we will discuss the challenges and progress in computational materials science.

Concerning the data-analytics, we note that the number of possible materials is practically infinite, but only 10 or 100 of them may be relevant for a certain science or engineering purpose. In simple words, in materials science and engineering, we are often looking for "needles in a hay stack". Fitting or machine-learning all data (i.e. the hay) with a single, global model may average away the specialties of the interesting minority (i.e. the needles). We will discuss methods that identify statistically-exceptional subgroups in a large amount of data, and how one can estimate the domains of applicability of machine-learning models. [3]

[1] FAIR stands for Findable, Accessible, Interoperable and Re-usable. *The FAIR Data Principles*; <https://www.force11.org/group/fairgroup/fairprinciples>.

[2] C. Draxl and M. Scheffler, *Big-Data-Driven Materials Science and its FAIR Data Infrastructure. Plenary Chapter in Handbook of Materials Modeling* (eds. S. Yip and W. Andreoni), Springer (2019). <https://arxiv.org/ftp/arxiv/papers/1904/1904.05859.pdf>.



[3] Ch. Sutton, M. Boley, L. M. Ghiringhelli, M. Rupp, J. Vreeken, M. Scheffler, *Domains of Applicability of Machine-Learning Models for Novel Materials Discovery*, to be published.

### ***Molecular simulation and machine learning as routes to exploring structure and phase behavior in atomic and molecular crystals***

**Mark E Tuckerman**

New York University College of Arts and Science, New York - USA

**Room Lausanne, 14:45**

**Monday 9**

Organic molecular crystals frequently exist in multiple forms known as polymorphs. Structural differences between crystal polymorphs can affect desired properties, such as bioavailability of active pharmaceutical formulations, lethality of pesticides, or electrical conductivity of organic semiconductors. Crystallization conditions can influence polymorph selection, making an experimentally driven hunt for polymorphs difficult. Such efforts are further complicated when polymorphs initially obtained under a particular experimental protocol “disappear” in favor of another polymorph in subsequent repetitions of the experiment. Consequently, theory and computational can potentially play a vital role in mapping the landscape of crystal polymorphism. Traditional crystal structure prediction methods face their own challenges, and therefore, new approaches are needed. In this talk, I will show, by leveraging concepts from statistical mechanics in combination with techniques of molecular simulation, traditional methods, and machine learning, that a new paradigm in crystal structure prediction may be emerging. Examples demonstrating prediction of structures of crystals, co-crystals, and phase transitions will be presented.

## **Contributed talks**

### ***Multiscale modeling of molecular diffusive transport phenomena: from microscopic details to macroscopic fluxes***

**Matteo Ceccarelli<sup>1</sup>**, Igor Bodrenko<sup>1</sup>, Stefan Milenkovic<sup>1</sup>, Silvia Acosta-Gutierrez<sup>2</sup>

<sup>1</sup>University of Cagliari, Cagliari - Italy

<sup>2</sup>University Central London, London - United Kingdom

**Room Blaricum, 17:15**

**Monday 9**

One of the challenges of modern medicine is the discovery of new antifectives to combat bacteria that are inherently resistant because of the low permeation of their cell membrane, namely those belonging to the Gram negative group. In this talk I'll summarize the effort of our group to combine diverse numerical simulation techniques for quantifying the transport of molecules through nanometric protein pores of the cell membranes. By applying a multi-scale approach, we moved from the microscopic details of molecules, namely the molecular properties related to specific chemical groups, to the macroscopic diffusive flux of molecules through nanopores, determined by the free energy landscape and the local diffusion coefficient. Once refined and automatized, such integrated bottom-up approach will allow the screening of virtual large databases in order to identify those molecular scaffold having optimal permeation through cell membranes. These permeating molecular scaffolds can be employed as input data of classical drug design techniques to add antifective properties and thus obtain new antibiotic compounds active against Gram negative bacteria.

### ***Autocatalytic patchy particles***

**Silvia Corezzi**, Cristiano De Michele, Francesco Sciortino

Dipartimento di Fisica e Geologia, Università di Perugia, Perugia - Italy

**Room Lyon, 17:15**

**Monday 9**

Autocatalysis, i.e., the speeding up of a reaction through the very same molecule which is produced, is common in chemistry, biophysics, and material science. Rate-equation-based approaches are often used to model the time dependence of products, but the key physical mechanisms behind the reaction cannot

be properly recognized. Here, building on previous studies on the subject [1-4], we develop a patchy particle model inspired by a bicomponent reactive mixture and endowed with adjustable autocatalytic ability [5]. Such a coarse-grained model captures all general features of an autocatalytic aggregation process that takes place under controlled and realistic conditions, including crowded environments. Simulation reveals that a full understanding of the kinetics involves an unexpected effect that eludes the chemistry of the reaction, and which is crucially related to the presence of an activation barrier. The resulting analytical description can be exported to real systems, as confirmed by experimental data on epoxy-amine polymerizations, solving a long-standing issue in their mechanistic description

[1] S. Corezzi, C. De Michele, E. Zaccarelli, D. Fioretto and F. Sciortino, *Soft Matter* **4**, 1173–1177 (2008).

[2] S. Corezzi, C. De Michele, E. Zaccarelli, P. Tartaglia and F. Sciortino, *J. Phys. Chem. B* **113**, 1233–1236 (2009).

[3] S. Corezzi, D. Fioretto, C. De Michele, E. Zaccarelli and F. Sciortino, *J. Phys. Chem. B* **114**, 3769–3775 (2010).

[4] S. Corezzi, D. Fioretto and F. Sciortino, *Soft Matter* **8**, 11207–11216 (2012).

[5] S. Corezzi, F. Sciortino and C. De Michele, *Nat. Commun.* **9**:2647 (2018).

### **Trajectory-based method for the study of ultrafast quantum dynamics**

**Lidice Cruz Rodríguez**<sup>1</sup>, Llinersy Uranga Piña<sup>2</sup>, Aliezer Martínez Mesa<sup>2</sup>, Christoph Meier<sup>1</sup>

<sup>1</sup>Laboratoire Collisions Agrégats Réactivité (LCAR), Institut de Recherche sur les Systèmes Atomiques et Moléculaires Complexes (IRSAMC), Toulouse - France

<sup>2</sup>Physics Faculty, Havana University, Havana - Cuba

**Room Lyon, 15:45**

**Monday 9**

The solution of the Schrodinger equation for high dimensional systems is one of the major challenges in contemporary physics, with implications in biology and chemistry. In spite of the development of sophisticated wave packet propagation methods, the description of the quantum dynamics of systems with more than a few atoms is in general not possible. In this sense, the development of trajectories based methods is an active field of research because of its favorable scaling properties with the system dimensionality. Within this context, a very attractive formalism is the hydrodynamic formulation of quantum mechanics where the Schrodinger equation is solved by propagating a bunch of trajectories under the combined action of quantum and classical forces. In this work, trajectory-based methods are employed to model quantum mechanical phenomena. Quantum trajectories are used as a tool to solve the hydrodynamic equations, without pre-computing the wave function. The advantage of the trajectories as an instrument to describe and interpret quantum dynamics are also exploited. Furthermore, approximate expressions for the quantum potential for one-dimensional and multidimensional model systems are derived by using different parametrizations of the density. The method is tested in some one-dimensional model potentials. Well-known quantum mechanical effects, such as zero-point energy, tunneling and scattering through potential barriers are captured in good agreement with standard wave packet propagation methods. Additionally, the same methodology is applied to describe the laser-matter interaction and both the ionization yield and the left-right asymmetry are evaluated as a function of the Carrier Envelope Phase. The multidimensional quantum potential is added to the classical interaction potential to obtain an effective potential, which is included in MD simulations in order to simulate the absorption spectra of atomic sodium embedded in argon and krypton matrices.

### **Modeling and simulating molecular kinetics as diffusion processes with Markovian switching**

**Mauricio J. Del Razo**, Frank Noé, Christoff Schütte, Manuel Dibak

Freie Universität Berlin, Berlin - Germany

**Room Lausanne, 17:15**

**Monday 9**

Molecular dynamics (MD) simulations can model the interactions between macromolecules with high spatiotemporal resolution but at a high computational cost. By coarse-graining MD into Markov state models (MSMs), it is now possible to obtain long time-scale behavior of some biomolecules and complexes constrained to small domains. Conversely, the interactions of many molecules in larger domains are better modeled by particle-based reaction-diffusion (PBRD) simulations, but they unfortunately lack molecular detail. Thus, coupling MSMs with PBRD simulations is highly desirable, as they could efficiently produce simulations at large time- and length-scales, while still conserving the

characteristic features of the interactions observed at atomic detail. In this work, we show this coupling can be achieved by coarse-graining MD simulations into a well-defined stochastic process that combines diffusion processes with Markov chains. We further derive and implement simulations of this newly developed coarse-grained model for simple systems, reducing the computational cost and allowing larger time- and length-scales without sacrificing molecular resolution.

### ***Emerging interactions through active alignment***

**Jure Dobnikar**<sup>1,2</sup>, Joan Codina<sup>1</sup>, Ignacio Pagonabarraga<sup>3</sup>

<sup>1</sup>Institute of Physics, CAS, Beijing - China

<sup>2</sup>University of Cambridge, Cambridge - United Kingdom

<sup>3</sup>CECAM Swiss Federal Institute of Technology (EPFL), Lausanne - Switzerland

**Room Lyon, 16:55**

**Monday 9**

Active suspensions are inherently out of equilibrium and can exhibit dynamic behavior not observed in their equilibrium counterparts. Basic building blocks for describing such systems are active particles that consume energy and convert it into motion. Upgrading previous models, where either propulsion or alignment was used as the active interaction, we here define a minimal coarse grained particle-based model for active systems that considers both self-propulsion and alignment of the particles. We argue that most living and synthetic active systems can be mapped onto this general model. The relevant dimensionless parameters of the model are the Péclet number,  $Pe$ , characterizing the propulsion intensity, and the reduced alignment strength,  $g$ . Within this general theoretical framework, we study the interaction mediated by the active suspension between passive inclusions. Upon varying the two parameters, we observe different qualitative regimes including short-range repulsion due to the capture of active particles between the inclusions; depletion-like attraction, and a double length scale interaction with repulsive and attractive short range part. Moreover, for sufficiently strong alignment, a long-range Casimir-like interaction is observed. We interpret the observed behavior in terms of competition between repulsive pressure due to the propulsion activity and attraction mediated by large-scale correlated dynamics due to the alignment torques.

### ***All-atom simulation of protein folding and direct validation against time-resolved spectroscopy experiments***

**Pietro Faccioli**<sup>1,2</sup>, Simone Orioli<sup>1</sup>, Giovanni Spagnolli<sup>1</sup>

<sup>1</sup>Trento University, Trento - Italy

<sup>2</sup>INFN, Trento - Italy

**Room Lausanne, 15:45**

**Monday 9**

The theoretical characterization of a protein folding process requires to overcome three main formidable challenges. First, the transition path ensemble for the folding reaction has to be accurately sampled. Next, the reactive trajectories generated by computer simulations have to be post-processed, in order to identify the kinetically relevant meta-stable states. Finally, this structural information needs to be translated into predictions for experimental observables, to enable experimental validation. In this talk, I discuss how these three major steps can be tackled by means of a specific combination of path integral-based enhanced sampling algorithms and approximation schemes [1], Renormalization Group-based statistical analyses [2], and excited-state quantum chemical calculations (required to connect molecular dynamics with time-resolved spectroscopy experiment) [3]. For illustration purposes, we report on applications of this scheme to predict the time-resolved single-molecule FRET and ensemble near-UV CD signals for the folding of several proteins consisting of several hundreds of amino-acids, using a realistic all-atom force field. Depending on time availability, we will also report about the application on the same scheme to simulate the interconversion between the cellular prion protein PrP-C into aggregates of the pathogenic scrapie isoform PrP-Sc [4].

[1] S. A. Beccara, T. Skrbic, R. Covino and P. Faccioli, *Proc. Natl. Acad. Sci. USA* **109** 2330 (2012) S. a Beccara, L. Fant and P. Faccioli, *Phys. Rev. Lett.* **114**, 098103 (2015). S. Orioli, S. a Beccara and P. Faccioli, *J. Chem. Phys.* **147**, 064108 (2017).

[2] S. Orioli and P. Faccioli, *J. Chem. Phys.* **145**, 124120 (2016).

[3] A. Ianeselli, S. Orioli, G. Spagnolli, P. Faccioli, L. Cupellini, S. Jurinovich, B. Mennucci, *J. Am. Chem. Soc.* **140**, 3674 (2018).

[4] G. Spagnolli et al, "Full Atomistic Model of Prion Structure and Conversion", to appear in *PLoS Pathogens* (2019).

### **Water splitting on hematite surfaces: insights from density-functional theory**

**Ralph Gebauer**

International Centre for Theoretical Physics, Trieste - Italy

**Room Orsay, 15:45**

**Monday 9**

The development of efficient ways to exploit the energy from the sun is an issue of major importance. Among possible solutions, the employment of solar energy to promote chemical reactions has the advantage of addressing the problems of harvesting, converting and storing energy at the same time. In this context, water splitting plays a central role both for direct hydrogen production and for the production of hydrocarbons. Therefore, great attention has been recently devoted to hydrogen production by means of photoelectrochemical (PEC) cells, via water splitting to molecular hydrogen and oxygen. The main challenge is to develop anode materials for these cells that can split water efficiently. Hematite ( $\alpha\text{-Fe}_2\text{O}_3$ ) has emerged as a highly interesting material for photoanodes. Abundant, stable, nontoxic, and, importantly, possessing an energy gap of 2.1 eV,  $\text{Fe}_2\text{O}_3$  has been intensively investigated in numerous experiments, often focusing on its photocatalytic properties. In this presentation, I will report recent numerical studies focusing on the oxidation of water on hematite (0001) surfaces. The water splitting reaction involves four consecutive proton coupled electron transfer steps. By means of density-function theory calculations, we investigate the role which the catalyst plays in those steps and how doping and defects of the iron oxide influence the photophysical activity of the anode.

### **Perpetual superhydrophobicity: drying from the nanoscale to the macroscale**

**Alberto Giacomello**

Sapienza Universita' di Roma, Roma - Italy

**Room Blaricum, 15:45**

**Monday 9**

Inspired by many biological examples, such as the lotus [1] or the *Salvinia molesta* [2] leaves, superhydrophobicity gained the attention of the scientific and engineering communities owing to the remarkable properties it confers to surfaces: self-cleaning, drag-reduction, anti-fouling, etc. One of the crucial limitations to the technological applicability of superhydrophobicity remains its fragility: once the superhydrophobic state, in which gaseous pockets are stabilized within surface roughness by capillary forces, experiences a transition to the fully wet Wenzel one, the relevant properties are lost and recovering them typically requires a significant external energy input. However, it has been recently shown by computer simulation that nanostructured surfaces can overcome this limitation promoting completely passive (self-)recovery of superhydrophobicity as a result of the combination of extreme confinement and hydrophobic environment, giving rise to "perpetual superhydrophobicity" [3,4]. This strategy is based on the thermodynamic elimination of the Wenzel state over a range of pressures of engineering interest: when pressure is decreased below a certain threshold the Wenzel state becomes unstable and superhydrophobicity is restored. In this work we identify the surface characteristics which favour this mechanism by leveraging macroscopic theory, classical density functional calculations, and rare-event molecular dynamics simulations [3,4,5]. Results show that surface cavities with extended hydrophobic surfaces and corners facilitate the destabilization of the Wenzel state; this drying mechanism, however, strongly depends on the scale of the cavities. Nanoscale hydrophobic cavities allow the thermodynamic destabilization of the confined liquid phase over an unexpectedly broad range of conditions, including pressures as large as 10 MPa and contact angles close to 90°. On the other hand, for cavities on the micron scale, such destabilization occurs only for much larger contact angles and close to liquid-vapor coexistence. These scale-dependent drying mechanisms are used to propose design criteria for hierarchical superhydrophobic surfaces capable of spontaneous self-recovery over a broad range of operating conditions. References

[1] W. Barthlott, W. and C. Neinhuis, *Planta*, **202**, page 1 (1997).

[2] M. Amabili, A. Giacomello, S. Meloni and , C.M. Casciola, *Advanced Materials Interfaces*, **2**, 1500248 (2015).

[3] A. Giacomello, L. Schimmele, S. Dietrich and M. Tassinkevych. *Soft Matter*, **12**, 8927 (2016).

[4] E. Lisi, M. Amabili, S. Meloni, A. Giacomello and C.M. Casciola, *ACS Nano*, **12**, page 359-367 (2017).

### ***Emergence of molecular friction in liquids: a memory-kernel perspective***

**Felix Höfling**, Arthur V. Straube, Bartosz G. Kowalik, Roland R. Netz

Freie Universität Berlin, Berlin - Germany

**Room Blaricum, 16:35**

**Monday 9**

Internal friction is a key ingredient for dynamic processes in molecular fluids: it limits diffusion, governs dissipation, and facilitates the relaxation towards equilibrium. At macroscopic scales, the steady motion of a tagged particle dragged through a viscous fluid obeys Stokes's friction law, which holds surprisingly well down to molecular scales. Microscopically, fluids are governed by Newton's equations of motion, which are conservative and time-reversible, and molecules do not experience friction. Using a memory kernel approach, which we link with a frequency-dependent friction via the generalised Langevin equation, we show how molecular friction emerges in liquids from the coupling of time scales. Here we report on an unusual, exponentially fast increase of the molecular friction in fluids as frequency is decreased from the microscopic limit, with the transition occurring near the Einstein frequency of the fluid. Our investigation was enabled by methodological advances in the harmonic analysis of correlation functions and in the extraction of memory kernels [1,2]. Most importantly, our model-free approach is based on the mean-square displacement of tracer particles as sole input and offers a framework suitable for the interpretation of experimental data from, e.g., microrheology. We corroborate our findings by extensive simulation data for water, a simple liquid, and a model glass former, each covering the full dynamic window from the molecular to the hydrodynamic regime. Analytical insight is obtained by solving a synthetic, yet physically meaningful example that possesses also a long-time tail.

[1] F. Höfling and T. Franosch, *Rep. Prog. Phys.* **76**, 046602 (2013).

[2] J. O. Daldrop, B. G. Kowalik, and R. R. Netz, *Phys. Rev. X* **7**, 041065 (2017).

### ***Dendrimer-like DNAs: simulation and experiment***

**Gerhard Kahl**<sup>1</sup>, Clemens Jochum<sup>1</sup>, Natasa Adzic<sup>2</sup>, Christos N. Likos<sup>2</sup>

<sup>1</sup>Institut für Theoretische Physik, Vienna University of Technology, Vienna - Austria

<sup>2</sup>Faculty of Physics, University of Vienna, Vienna - Austria

**Room Lyon, 16:35**

**Monday 9**

Dendrimers are synthetic macromolecules, characterized by a highly branched and regular internal architecture. Recently, dendrimer-like DNAs (DL-DNAs) were synthesized via enzymatic ligation of Y-shaped DNA building blocks. These charged dendrimers represent a novel macromolecular aggregate, which holds high promise in bringing about targeted self-assembly of soft-matter systems in the bulk and at interfaces. We present a joint simulational-experimental study of these novel macromolecules. Based on a bead-spring model for the DL-DNAs (of varying generation numbers) we perform large-scale simulations to determine the equilibrium properties and the conformational characteristics of these macromolecules. The obtained results are compared to light scattering experiments [1]. The simulation data provide a broad variety of additional information about the internal molecular structure of DL-DNAs by varying the generation number and the salinity of the solvent. In an effort to simulate concentrated solutions of DL-DNAs, we extract an effective, coarse-grained potential, based on Widom's particle-insertion method. With this potential at hand, we investigate the bulk behaviour of DL-DNAs. These findings are essential to investigate if these macromolecules are a viable candidate for the experimental realization of cluster crystals with multiple site occupancy in the bulk [2]. The study of these charged dendrimer systems represents a relevant field of research in the area of soft matter due to their potential role for various interdisciplinary applications, ranging from molecular cages for drug delivery to the development of dendrimer- and dendron-based ultra-thin films in the area of nanotechnology [3].

[1] C. Jochum, N. Adzic, E. Stiakakis, T.L. Derrien, D. Luo, G. Kahl, and C.N. Likos, *Nanoscale* **11**, 1604 (2019).

[2] B. Mladek, D. Gottwald, M. Neumann, G. Kahl, C. N. Likos, *Phys. Rev. Lett.* **96**, 45701 (2006).

[3] C. Lee, J. MacKay, J. Frechet, F. Szoka, *Nat. Biotechnol.* **23**, 1517 (2005).

**Simulation of NMR spectra for molecules or ions diffusing in porous disordered carbons**  
Céline Merlet<sup>1</sup>, Dimitrios Kilymis<sup>1</sup>, Albert P. Bartók<sup>2</sup>, Chris J. Pickard<sup>3</sup>, Daan Frenkel<sup>4</sup>, Clare P. Grey<sup>4</sup>

<sup>1</sup>CIRIMAT, Université de Toulouse, CNRS, Toulouse - France

<sup>2</sup>STFC, Rutherford Appleton Laboratory, Didcot –United Kingdom

<sup>3</sup>Department of Materials Science and Metallurgy, University of Cambridge, Cambridge - United Kingdom

<sup>4</sup>Department of Chemistry, University of Cambridge, Cambridge - United Kingdom

**Room Blaricum, 16:55**

**Monday 9**

Local fluid structure and ion transport in porous materials are relevant for a number of applications including energy conversion and storage, heterogeneous catalysis and drug delivery. In all these cases, the performance of the systems is highly related to the specific properties of the fluid under confinement. In order to understand fundamentally the macroscopic properties of such systems, it is essential to characterise finely the porous materials used and the structural and dynamic properties of the fluid adsorbed. Nuclear Magnetic Resonance (NMR) has proven to be a very useful technique for the study of these materials, in particular when combined with DFT (Density Functional Theory) calculations which provide key information to assign and interpret NMR spectra. Nevertheless, in some cases, DFT calculations are not sufficient as dynamical processes can lead to complex NMR spectra where individual environments are hard to identify. In this talk I will focus on porous carbons and report on how lattice simulations can be used to model ion diffusion in carbon particles and predict the NMR spectra of such ions [1]. The lattice model includes information from molecular dynamics simulations such as the free-energy profile for ionic adsorption, and density-functional theory calculations are used to predict the NMR chemical shift of the adsorbed ions. This allowed us to estimate the size of aromatic domains in several disordered porous carbon materials [2]. To go beyond this domain size estimation, there is a need for a better description of the chemical shift of adsorbed species. Indeed, in the past, chemical shifts calculations were simply done on small planar aromatic molecules. I will show that for small aromatic molecules, not necessary planar, a perturbation approach applied on a simple Hückel Hamiltonian can give accurate results. A study is ongoing to improve this model in order to make it suitable for periodic disordered carbons.

[1] C. Merlet, A. C. Forse, J. M Griffin, D. Frenkel and C. P. Grey, *J. Chem. Phys.*, **142**, 094701 (2015).

[2] A. C. Forse, C. Merlet, P. K. Allan, E. K. Humphreys, J. M Griffin, M. Aslan, M. Zeiger, V. Presser, Y. Gogotsi and C. P. Grey, *Chem. Mater.*, **27**, 6848 (2015).

**Singlet fission molecules among known compounds: finding a few needles in a haystack**  
Daniele Padula<sup>1</sup>, Alessandro Troisi<sup>2</sup>

<sup>1</sup>Laboratory for Functional Polymers, Swiss Federal Laboratories for Materials Science and Technology, Empa, Dübendorf, Switzerland

<sup>2</sup>Department of Chemistry, University of Liverpool, Liverpool - United Kingdom

**Room Orsay, 16:55**

**Monday 9**

A large set of candidates for singlet fission, one of the most promising processes able to improve the efficiency of solar cells, are identified by screening a database of known molecular materials. The screening was carried out through a procedure exploiting quantum chemical calculations of excited state energies, carefully calibrated against a substantial set of experimental data. We identified ~200 potential singlet fission molecules, the vast majority of which were not known as singlet fission materials. The molecules identified could be grouped into chemical families, enabling the design of further singlet fission materials using the hits as lead compounds for further exploration. Many of the discovered materials do not follow the current design rules used to develop singlet fission materials, illustrating at the same time the power of the screening approach and the need for developing new design principles.

### ***Neural network based path collective variables for enhanced sampling of phase transformations***

**Jutta Rogal**<sup>1</sup>, Elia Schneider<sup>2</sup>, Mark Tuckerman<sup>2</sup>

<sup>1</sup>Ruhr-Universität Bochum, Bochum - Germany

<sup>2</sup>New York University, New York - USA

**Room Orsay, 16:35**

**Monday 9**

Atomistic simulations of solid-solid phase transformations provide insight into the microscopic processes that govern the kinetics and mechanisms of the transition. Understanding these processes is of considerable technological importance in a wide range of materials such as metals and alloys, minerals, or molecular crystals. A particular challenge in the sampling of solid-solid phase transformations is the involved timescales that often reach far beyond what is accessible with classical molecular dynamics. To achieve an efficient sampling of the underlying high dimensional phase space with often complex features typically requires the application of enhanced simulation approaches. Here, we combine machine learning techniques with enhanced sampling algorithms to explore structural phase transformations. Specifically, we employ a neural network based identification of local structural environments that are then used to construct global structural classifiers. Within the space of global classifiers a one-dimensional path collective variable is defined that is subsequently used in the enhanced sampling. We illustrate our approach by exploring the complex migration of a phase boundary during the solid-solid transformation between the body-centred cubic and topologically closed packed A15 phase in molybdenum. Within the simulations we were able to observe, for the first time, the growth of both the BCC and A15 phase, and estimate the free energy profile along the transformation. The approach presented here is generally applicable, and the efficient sampling of the phase space along the one-dimensional path collective variable can facilitate the study of structural transformations in complex condensed matter systems, such as molecular crystals.

### ***Modeling crystal nucleation with the string method***

**Erik E. Santiso**

North Carolina State University, Raleigh, - USA

**Room Lausanne, 16:35**

**Monday 9**

Crystal nucleation is critical to many applications in the food, chemical, construction, and pharmaceutical industries, among others. However, crystal nucleation is challenging to study both experimentally and theoretically due to the time and length scales involved, and the diversity of systems and crystalline structures of practical relevance. In this talk we will present an overview of our work using the string method in collective variables to study crystal nucleation of organic molecules, including variations of the method capable of studying crystallization in open-system ensembles. We also highlight some of the challenges and potential directions to further study this problem using molecular simulation.

### ***Machine learning potentials for molecular liquids***

**Max Veit**<sup>1</sup>, Sandeep Kumar Jain<sup>2</sup>, Satyanarayana Bonakala<sup>2</sup>, Indranil Rudra<sup>2</sup>, Detlef Hohl<sup>3</sup>, Gábor Csányi<sup>4</sup>

<sup>1</sup> Swiss Federal Institute of Technology (EPFL), Lausanne - Switzerland

<sup>2</sup>Shell Technology Centre Bangalore, Bengaluru, Karnataka - India

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**Room Orsay, 17:15**

**Monday 9**

The reliable prediction of the macroscopic properties of molecular liquids requires potential energy surface (PES) models that are not only accurate, but computationally efficient enough to handle large systems and reach long time scales typically inaccessible to explicit quantum-mechanical methods. This work introduces a new approach to the systematic approximation of the first-principles PES of a molecular liquid using the GAP machine learning method [1]. By applying machine learning to separately approximate each physical component of the interaction energy in a full many-body framework and with high and controllable accuracy, we can simulate the liquid accurately across a wide range of temperatures

and pressures (with the inclusion of quantum nuclear effects) while gaining physical insight into the inner workings of the fluid. Following the recent success of this approach on predicting the equation of state of compressed fluid methane [2], I will show how this approach can be extended to other molecular liquids with the help of emerging techniques in machine learning potential development, and how these ideas can be applied to other important molecular materials.

[1] A. Bartók, M. Payne, R. Kondor, and G. Csányi, *Phys. Rev. Lett.* **104**, 136403 (2010).

[2] M. Veit, S. K. Jain, S. Bonakala, I. Rudra, D. Hohl, and G. Csányi, *JCTC* **15**, 2574 (2019).

### **Learning clustered representation for complex free energy landscapes**

**Jun Zhang**<sup>1</sup>, Yao-Kun Lei<sup>1</sup>, Xing Che<sup>2</sup>, Zhen Zhang<sup>1</sup>, Yi Isaac Yang<sup>3</sup>, Yi-Qin Gao<sup>1</sup>

<sup>1</sup>College of chemistry and molecular engineering, Peking University, Beijing - China

<sup>2</sup>Kasha Laboratory of Biophysics, Florida State University, Tallahassee - USA

<sup>3</sup>Department of Chemistry and Applied Biosciences, ETH Zürich, Zürich - Switzerland

Room Lausanne, 16:55

Monday 9

We first analyzed the inductive bias underlying the data scattered across complex free energy landscapes (FEL), and exploited it to train deep neural networks which yield reduced and clustered representation for the FEL. Our parametric method, called Information Distilling of Metastability (IDioM), is end-to-end differentiable thus scalable to ultra-large dataset. IDioM is also a clustering algorithm and is able to cluster the samples in the meantime of reducing the dimensions. Besides, as an unsupervised learning method, IDioM is superior to many existing dimensionality reduction and clustering methods in that it neither requires a cherry-picked distance metric nor the ground-true number of clusters. We show that the reduced representation learned by IDioM outperforms many popular methods and is amenable for downstream tasks such as kinetic modeling because IDioM partitions the FEL into well-defined metastable states.



**Plenary speakers*****Advances in interatomic potentials for materials*****Gabor Csanyi**

Cambridge University, Cambridge - United Kingdom

Room Lausanne, 17:25

Tuesday 10

Modelling the atomic scale properties of materials is one of the success stories over the past four decades. Increasingly complex functional forms, from pair potentials to embedded atom models and bond order potentials, allowed the quantitative description of different crystal structures, point and line defects, surfaces, shedding light on many elementary processes governing failure, phase stability, etc. The accuracy with which these models describe the potential energy surface corresponding to the electronic ground state has not changed much over the decades and is rather limited. The success is thus largely empirical in nature - and follows from the sophistication of the modeller and the judicious compromises made in order to solve specific problems. The parallel developments in electronic structure theory on the other hand provided exquisite quantitative agreement with experiments. I will report on recent work of a growing community, who have managed to bring these two worlds together, and construct extremely accurate functional representations of the interatomic potential. These developments rely on a very large amount of highly accurate electronic structure data, on non-parametric function fitting, and on sophisticated representation theory that brings with it guarantees of completeness and convergence.

***Light-activated matter: from photo-electrochemical cells to optogenetics and quantum information systems*****Giulia Galli**

University of Chicago, Chicago - USA

Room Lausanne, 09:00

Tuesday 10

We discuss first principles, computational methods and strategies to predict light-activated processes in sustainable materials for use in photoelectrochemical cells, optogenetic and quantum information systems.

***Fast irreversible markov chains in statistical physics*****Werner Krauth**

Ecole Normale Supérieure, Laboratoire de Physique, Paris - France

Room Lausanne, 14:15

Tuesday 10

The Monte Carlo method is an outstanding computational tool in science. Since its origins, it has relied on the detailed-balance condition (that is, the absence of flows in equilibrium) to map general computational problems onto equilibrium-statistical-physics systems.

Irreversible Markov chains violate the detailed-balance condition. They realize the equilibrium Boltzmann distribution as a steady state with non-vanishing flows. For one-dimensional particle models we have proven rigorously that local algorithms reach equilibrium on much faster time scales than the reversible algorithms that satisfy detailed balance. The event-chain Monte Carlo algorithm (ECMC) generalizes these irreversible Markov chains to higher dimensions. It relies on a factorized Metropolis filter which is based on a consensus rule rather than on an energy criterion.

As applications I will discuss the solution of the famous two-dimensional melting problem for hard disks and related systems, where we showed, using ECMC, that hard disks melt neither following the Kosterlitz-Thouless-Halperin-Nelson-Young prescription nor the alternative first-order liquid-solid scenario. I will also present the use of ECMC for the general classical all-atom N-body problem. Here, the Boltzmann

distribution  $\exp(-\beta E)$  is sampled (without any discretization or truncation error) but the potential energy  $E$  remains unknown. This is of great interest in the Coulomb problem, where  $E$  or its derivatives, the forces, are hard to compute. Our recent JeLLyFysh open-source Python application implements ECMC for models from hard spheres to three-dimensional water systems. I will finish by discussing its features, that closely mirror the mathematical formulation of ECMC, and by presenting its future challenges.

## **Invited speakers**

### ***Self-folding kirigami at the microscale***

**Nuno Araujo**

University of Lisbon, Lisbon - Portugal

**Room Lyon, 11:30**

**Tuesday 10**

Three-dimensional shells can be synthesized from the spontaneous self-folding of two-dimensional templates of interconnected panels, called nets. However, some nets are more likely to self-fold into the desired shell under random movements. The optimal nets are the ones that maximize the number of vertex connections, i.e., vertices that have only two of its faces cut away from each other in the net. Previous methods for finding such nets are based on random search and thus do not guarantee the optimal solution. We proposed a deterministic procedure [1]. Our method allows not only to design the self-assembly of much larger shell structures but also to apply additional design criteria, as a complete catalog of the nets with the maximum number of vertex connections is obtained.

[1] N. A. M. Araújo, R. A. da Costa, S. N. Dorogovtsev, J. F. F. Mendes, *Phys. Rev. Lett.* **120**, 188001 (2018).

### ***Equilibrium simulations of supercooled liquids beyond laboratory timescales***

**Ludovic Berthier**

University of Montpellier, Montpellier - France

**Room Blaricum, 11:30**

**Tuesday 10**

Computer simulations give unique insights into the microscopic behavior of disordered and amorphous materials, but their typical timescales are orders of magnitude shorter than the experimentally relevant ones. In particular, simulations of supercooled liquids performed with standard techniques cover at most 4-5 decades of viscous slowing down, far behind the 13 decades commonly accessible in experimental studies. Recently, we have closed this enormous gap for a class of realistic models of liquids, which we can successfully equilibrate beyond laboratory time scales by means of a swap Monte Carlo algorithm. For some models, we achieve over 10 orders of magnitude speedup in equilibration timescale. This exciting numerical advance allows us to address some outstanding questions concerning the formation and properties of glasses in a dynamical range that remains inaccessible in experiments, such as the relevance of an entropy crisis, the kinetics of ultrastable glasses, and the rheology of realistic glassy materials.

### ***Emergence of knots in intrinsically disordered proteins***

**Marek Cieplak**

Polish Academy of Science, Warsaw - Poland

**Room Lyon, 15:35**

**Tuesday 10**

Transient knotted structures are expected to arise during the erratic evolution of intrinsically disordered peptide chains. We show that this is indeed the case for sufficiently long polyglutamine tracts and  $\alpha$ -synuclein. The polyglutamine tracts are fused within huntingtin protein that is associated with the Huntington neurodegenerative disease. We show that the presence of knots in the tracts hinders and sometimes even jams proteasomal translocation, especially when the knots are deep. The knots in polyglutamine may form in tracts exceeding about 40 residues. This fact should explain the existence of

a similarly sized length threshold above which there is an experimentally observed toxicity at the monomeric level. We also discuss emergence of knots in  $\alpha$ -synuclein. We show that these knots are shallow and they multiply form and disappear in a time interval of about 3  $\mu$ s, as inferred from an all-atom explicit-solvent 30  $\mu$ s trajectory. We discuss conformational biases that take place in  $\alpha$ -synuclein and establishment of contact during aggregation of two chains of this protein. We then discuss several aspects of dynamics of knotted structured proteins as assessed within a Go-like model. In particular, we argue that folding under the nascent conditions is essential to fold to a structure that is deeply knotted.

*In collaboration with: M. Chwastyk, Ł. Mioduszeowski, A. Gomez-Sicilia, M. Carrion-Vazquez, P. Robustelli, and Y. Zhao.*

### ***The physics of smarter and more sustainable cements***

**Emanuela Del Gado**

Georgetown University, Washington - USA

**Room Lausanne, 15:05**

**Tuesday 10**

Concrete and cement are the foundation of our global infrastructure and have a key role in the growth which is required to support a world population projected to surge past 9 billion by mid-century, with more than 70% of it living in urban areas. More than 20 billion tons of concrete are produced every year, more than any other material on Earth, such that concrete production is responsible for nearly 10% of the whole anthropogenic production of CO<sub>2</sub>. Reducing CO<sub>2</sub> emissions for concrete production, designing and optimizing material performances, resilience and durability is hence crucial to a sustainable growth and to meet Green House Emissions reduction goals. Strength and other mechanical properties of concrete rely on cement (its main binding agent) and the control of the formation/gelation of calcium-silicate-hydrates (C-S-H). Lack of scientific insight into the structure and mechanics of C-S-H is a major obstacle to optimizing material performances and developing greener formulations of concrete. In recent years electron microscopy imaging, nano-indentation tests, X-rays and neutron scattering, NMR analysis, and atomistic simulations have elucidated several structural and mechanical features concentrated within a few nanometers. A potential breakthrough has been combining such experimental insights with novel fundamental understanding gained through modeling and numerical simulations, which use statistical and condensed matter physics approaches to tackle the structural and mechanical complexity of the material over critical lengthscales. These achievements provide novel opportunities to transform cement production and use.

### ***The role of dynamin twist in membrane fission***

**Markus Deserno<sup>1</sup>, Martina Pannuzzo<sup>2</sup>, Zachary A. McDargh<sup>3</sup>**

<sup>1</sup>Carnegie Mellon University, Pittsburgh -USA

<sup>2</sup>Istituto Italiano di Tecnologia, Genova - Italy

<sup>3</sup>Dept. of Chemical Engineering, Columbia University, New York - USA

**Room Blaricum, 11:00**

**Tuesday 10**

The final step of many biological membrane fission events involves the GTPase dynamin, which assembles into a helical filament around the neck of a nascent vesicle and somehow severs this remaining connection. But despite about two decades of research, the actual physical processes that lead to fission are still a matter of debate. Dynamin's action occurs on the scale of a few tens of nanometers over just a few milliseconds, which is too small and fast for many experimental techniques, but too large and slow for atomistic simulations. In this talk I explore this problem using both continuum theory and coarse-grained simulations specifically designed to capture the interplay of geometry and elasticity. I show that within reasonable experimental limits, the two widely discussed conformational changes of shrinking the radius or increasing the pitch of a dynamin helix are insufficient to trigger fission. However, a third change, reminiscent of an effective twist of the filament, turns out to efficiently drive the neck into the hemifission state. This motion mirrors the experimentally observed asymmetric tilting of dynamin's PH-domains. Following the retraction of the substrate, the remaining dynamin coat can unbind, and the tensile force in the connecting micellar string draws the almost severed membranes together one more time, until bilayer contact catalyzes the scission of the micelle.

### ***Birth of a binary crystal nucleus of hard spheres***

**Marjolein Dijkstra**

University of Utrecht, Utrecht - Netherlands

**Room Blaricum, 15:35**

**Tuesday 10**

Photonic crystals have numerous applications ranging from optical waveguides to ultra-fast optical computers. One way to fabricate photonic crystals is via colloidal self-assembly. The diamond and pyrochlore lattices are the most sought-after structures because of their wide photonic bandgaps. However, these open lattices are difficult to self-assemble as they are mechanically unstable for simple repulsive colloids. To find away around, one can self-assemble both the diamond and pyrochlore structure in a close-packed  $\text{MgCu}_2$  Laves phase (LP) from a binary colloidal dispersion. By removing the large/small spheres (Mg/Cu) from the  $\text{MgCu}_2$  phase, one can readily obtain the diamond/pyrochlore lattice. The three LPs,  $\text{MgCu}_2$ ,  $\text{MgNi}_2$ ,  $\text{MgZn}_2$ , can be assembled in a binary hard-sphere mixture. As all three LPs pack with the same density, the free-energy difference between them is very small, with  $\text{MgZn}_2$  the most stable phase. Despite this, LPs have never been seen to crystallize in such a mixture in simulations and experiments on colloids with sizes that open up a photonic bandgap in the visible. Using simulations we show that binary hard-sphere mixtures get arrested into a glass, thereby preventing LP nucleation, due to the presence of fivefold clusters in the supersaturated fluid. Our results reveal that by introducing softness in the interparticle potential, one can postpone the glass transition by reducing the concentration of fivefold structures in the fluid phase in such a way that crystal nucleation can proceed without interruption. This finding enabled us to show the first ever instance of spontaneous LP nucleation in nearly hard spheres. Unfortunately the most stable phase in bulk is the  $\text{MgZn}_2$  phase, and not the aimed  $\text{MgCu}_2$ . Here, we show by simulations that up to one hundred thousands of these particles in a spherical confinement spontaneously crystallize into a *3D icosahedral binary quasicrystal* consisting of twenty tetrahedral domains with  $\text{MgCu}_2$ -like symmetry. Finally, we investigate one of the many open questions on icosahedral quasicrystals --- Can we watch the birth of an icosahedral quasicrystal? Our simulations show for the first time the birth of an icosahedral quasicrystal of hard spheres.

### ***Computer simulations of biological systems: proteins, RNA, and membranes***

**Ron Elber**

The University of Texas at Austin, Austin - USA

**Room Lausanne, 11:00**

**Tuesday 10**

I discuss atomically detailed simulations of biological systems using the methodologies of Molecular Dynamics and Milestoning. These atomically detailed simulations are challenging due to the extremely wide range of timescales relevant to their functions. We illustrate how the use of short molecular dynamics trajectories can facilitate the study of longtime events. We consider conformational transition in molecular machines; the folding of a model RNA molecule with a helix-junction-helix for a secondary structure, and translocation events of charged species through biological membranes.

### ***The physics of crumpling and folding of untangled polymers and chromosomes***

**Ralf Everaers**

École normale supérieure de Lyon, Lyon - France

**Room Lyon, 15:05**

**Tuesday 10**

The conformational statistics of ring polymers in melts or dense solutions is strongly affected by their quenched microscopic topological state. The effect is particularly strong for untangled (i.e. non-concatenated and unknotted) rings, which are known to crumple and segregate. In particular there is evidence, that sufficiently long (e.g. human) chromosomes fall into this class in between cell divisions, because the reptation time necessary to entangle with each other far exceeds biologically relevant time scales. We have used extensive simulation studies to compare the predictions of polymers models to biological data and to elucidate the physics underlying the behavior crumpled polymers. With respect to the (biological) relevance of crumpled polymers, I will present comparisons to HiC experiments in a study of the large scale chromosome folding in *Drosophila* nuclei during the course of development. We find

that territory formation is fully described by the idea of topologically constrained relaxation of decondensing metaphase chromosomes. The characteristic signature of Rabl territories due to the memory of quasi-nematic chromosome alignment is visible during early stages of development, but disappears in late embryo nuclei. Compartmentalization of centromeric heterochromatin is well accounted for by co-polymer models with like-like attraction between hetero- and eu-chromatin. The additional distinction of a small number of epigenetic states allows to reasonably well predict the formation of (and interaction between) TADs. Concerning the underlying physics, I will focus on our “Feynman test” of theoretical ideas for the large scale ring structure: can we convert them into algorithms for constructing (nearly) equilibrated ring melt samples? We find that most structural observables are quantitatively reproduced by two different construction schemes: hierarchical crumpling and ring melts derived from the analogy to melts of randomly branching polymers. However, the latter fail the “Feynman test” with respect to the magnetic radius, which we have defined based on an analogy to magnetostatics. While the magnetic radius is expected to vanish for double-folded structures, the observed values provide a simple and computationally convenient measure of the presence of a non-negligible amount of local loop opening in crumpled rings.

A. Rosa, R. Everaers, *PLoS Computational Biology* **4**, e1000153 (2008).

A. Rosa, R. Everaers *Phys. Rev. Lett.* **112**, 118302 (2014).

R.D. Schram, A. Rosa, R. Everaers, *Soft Matter* **15**, 2418-2429 (2019).

P. Carrivain, Y. Ogiyama, C. Vaillant, G. Papadopoulos, D. Jost, G. Cavalli, R. Everaers, *in preparation*.

### **Active filaments, membranes, and cells**

**Gerhard Gompper**, Thorsten Auth, Jens Elgeti, Roland G. Winkler  
Forschungszentrum Julich, Julich - Germany

Room Lausanne, 15:35

Tuesday 10

Active matter exhibits a wealth of emerging non-equilibrium behaviors [1]. A paradigmatic example is the interior of cells, where active components, such as the cytoskeleton, are responsible for its structural organization and the dynamics of the various components. Of particular interest are the properties of active polymers and filaments [2]. The intimate coupling of active forces, thermal noise, hydrodynamic interactions, and polymer connectivity implies the emergence of novel structural and dynamical features. Different propulsion mechanisms capture the physics of a variety of systems, such as chains of active Brownian particles [3], polar filaments propelled along their contours [4,5], or cytoskeletal filaments propelled by motor bundles [6]. This leads to interesting single-particle behavior, such as a softening of a semiflexible filament of active Brownian particles at intermediate levels of activity [3], or a sperm-like beating motion of a filament pushing a load. At high polymer densities in two dimensions, collective dynamics characterized by active turbulence is observed [5].

Closed polymer rings (in two-dimensions) can be considered as a model of cell membranes. Here, active components lead to enhanced fluctuations [7] and an intimate coupling of propulsion forces, membrane deformability, cell shape, and cell sensing and reactivity [8,9].

[1] J. Elgeti, R.G. Winkler, and G. Gompper, *Rep. Prog. Phys.* **78**, 056601 (2015).

[2] R.G. Winkler, J. Elgeti, and G. Gompper, *J. Phys. Soc. Japan* **86**, 101014 (2017).

[3] T. Eisenstecken, G. Gompper, and R.G. Winkler, *J. Chem. Phys.* **146**, 154903 (2017).

[4] R.E. Isele-Holder, J. Elgeti, and G. Gompper, *Soft Matter* **11**, 7181 (2015).

[5] O. Duman, R.E. Isele-Holder, J. Elgeti, and G. Gompper, *Soft Matter* **14**, 4483 (2018).

[6] A. Ravichandran, O. Duman, M. Hoore, G. Saggiarato, G.A. Vliegthart, T. Auth, and G. Gompper, *eLife* **8**, e39694 (2019).

[7] S.M. Mousavi, G. Gompper, and R.G. Winkler, *J. Chem. Phys.* **150**, 064913 (2019).

[8] C. Abaurrea Velasco, S.D. Ghahnaviyeh, H.N. Pishkenari, T. Auth, and G. Gompper, *Soft Matter* **13**, 5865 (2017).

[9] C. Abaurrea Velasco, T. Auth, and G. Gompper, *arXiv:1812.09932* (2018).

***Simulations of structural transformations in ceramide phases from fitting structures to CEMOVIS image data***

**Erik Lindahl**<sup>1</sup>, Magnus Lundborg<sup>2</sup>, Christian Wennberg<sup>2</sup>, Ali Narangifard<sup>3</sup>, Lars Norlén<sup>4</sup>,

<sup>1</sup>Stockholm University, Stockholm - Sweden

<sup>2</sup>ERCO Pharma, Solna - Sweden

<sup>3</sup>Karolinska Institutet, Solna - Sweden

<sup>4</sup>Karolinska Hospital, Solna - Sweden

**Room Lausanne, 10:30**

**Tuesday 10**

The stratum corneum is the outermost layer of human skin and primary barrier towards the environment. The main component is stacked layers of saturated long-chain ceramides, free fatty acids and cholesterol, but we do not yet know the molecular structure or formation details. Here, I will present our work on new methods to fit models to low-resolution cryo-EM microscopy vitreous section (CEMOVIS) data, in particular by generating molecular models and using cryo-EM simulation to generate electron diffraction micrographs that can be compared directly to experimental data, and iteratively use these to improve the models. This has enabled us to create a number of alternative models, compare how they fit existing experimental data, and also use coarse-grained simulations to understand the formation process where cubic phases turn into bilayers depending on the lipid composition. These types of models can be highly useful tools for understanding the barrier properties, and we are currently combining it with free energy calculations to explore rapid prediction of permeation properties from CEMOVIS-derived models, which could have important applications in developing new generations of skin-permeating drugs.

***Biophysical principles of transcription-driven chromosome organisation***

**Davide Marenduzzo**

University of Edinburgh, Edinburgh- United Kingdom

**Room Lausanne, 11:30**

**Tuesday 10**

Current models for the folding of the human genome see a hierarchy stretching down from chromosome territories, through A/B compartments and TADs (topologically-associating domains), to contact domains stabilized by cohesin and CTCF. However, molecular mechanisms underlying this folding, and the way folding affects transcriptional activity, remain still to some extent obscure. I will describe some biophysical principles for genome organisation arising from molecular dynamics simulations of a simple 3D polymer model, in which chromatin fibres and whole chromosomes interact with bivalent or multivalent chromatin-binding proteins (such as transcription factors binding to their cognate sites).

Chromatin binding mediates cooperative interactions between proteins which naturally leads to the creation of clusters. Such clusters are strikingly similar to some of the "nuclear bodies" and transcription factories found inside the nucleus of eukaryotic organisms. The model also provides potential solutions to some puzzles concerning the actions of enhancers, super-enhancers, and expression quantitative trait loci (eQTLs). If time permits, I will discuss how the model can be used to study the dynamics of chromatin transcription, and to quantitatively predict the effect of 3D structure on transcriptional activity in human cells.

***Modeling approaches for soft glassy rheology***

**Kirsten Martens**

University Grenoble Alpes, Saint-Martin-d'Hères - France

**Room Lyon, 11:00**

**Tuesday 10**

In this talk I will discuss modeling approaches for the athermal rheology of soft glasses. Starting from observation on the microscopic scale to derive elasto-plastic descriptions on a mesoscopic scale, I will show how this modeling strategy can help to single out specific ingredients in the microscopic dynamics to explain emergent macroscopic features in athermally driven soft glasses. The coarse grained descriptions can help us to understand better a series of phenomena, such as creep, transient and permanent shear banding, avalanche dynamics in the low shear rate limit and a novel out-of-equilibrium critical point at finite driving rate.

### **Structure formation in copolymer materials after sudden quenches**

**Marcus Müller**

Georg-August-Universität Göttingen, Göttingen - Germany

**Room Blaricum, 10:30**

**Tuesday 10**

The free-energy landscape of multi-component polymer systems is rough and characterized by multiple metastable minima. It depends on thermodynamical control parameters such as temperature or pressure/strain. Polymer materials are ideal model systems to explore how one can deterministically direct the assembly of multicomponent polymer systems by sudden quenches of the thermodynamic control parameters because the time scales of collective structure changes are large and an accurate mean-field description for equilibrium properties is available. Diblock copolymers play an important role for the fabrication of battery materials and fuel cells. Using large-scale simulations we investigate the non-equilibrium morphology of diblock copolymers after a quench from the disordered phase. After the initial spinodal self-assembly, the morphology evolution becomes trapped in one of many metastable states. First, by combining particle-based simulation and numerical self-consistent field theory we define "inherent morphologies" that assign to each configuration of this self-assembling liquid a free-energy basin. Second, we explore the large-scale transport properties of these these experimentally relevant, non-equilibrium states. We observe three-dimensionally percolating network structures already for small volume fractions of the conducting component and find that the conductivity of the non-equilibrium structures is significantly smaller than that of the corresponding equilibrium phases. Moreover, we identify large but finite-sized, fractal-like structures inside the morphology, which influence the transport properties.

### **Calculation of electronic spectra using auxiliary systems and model results**

**Lucia Reining**

Ecole Polytechnique, Palaiseau - France

**Room Orsay, 10:30**

**Tuesday 10**

One of the most powerful ideas to approach the many-body problem is to build a simplified auxiliary system, meant to provide certain observables, but not others. The Kohn-Sham system in Density Functional Theory (DFT), which yields the density, is the most prominent example. Since it is difficult to design such an auxiliary system, sometimes results from models, such as the homogeneous electron gas, are used in an approximate way, for example in the Local Density Approximation. We generalize the concepts of an auxiliary system, and of the use of model systems from which information is imported [1], in order to set up a framework that allows us to calculate any observable, including electronic spectra, in an in principle exact way. We discuss promising strategies for approximations, and show that results are often superior to straightforward perturbation theory. Illustrations will include problems of DFT as well as correlation effects in inelastic x-ray scattering spectroscopies [2] and photoemission.

[1] M. Vanzini, A. Aouina, M. Panholzer, M. Gatti and L. Reining, *arXiv:1903.07930*.

[2] M. Panholzer, M. Gatti, and L. Reining, *Phys. Rev. Lett.* **120**, 166402 (2018).

### **Nuclear quantum effects in first-principles materials simulations**

**Mariana Rossi**

Fritz Haber Institute of the Max Planck Society, Berlin, Germany

**Room Orsay, 15:05**

**Tuesday 10**

In this talk, I will discuss techniques based on path integral molecular dynamics to include temperature and nuclear quantum effects in electronic-structure calculations. We achieve an all-atom, all-electron quantum description of these systems by developing methodology and tools that allow joining density-functional-theory calculations to path-integral methods, and which we implement in collaborative program packages available to the community [1,2]. As I will discuss, this combination allowed us solve questions related to level broadening in semiconductor interfaces, isotope effects in electron-phonon coupling and apparently contradictory observations in hydrogen-transfer reactions [3-5].

- [1] V. Blum, R. Gehrke, F. Hanke, P. Havu, V. Havu, X. Ren, K. Reuter, and M. Scheffler: *Ab initio molecular simulations with numeric atom-centered orbitals*. *Computer Physics Communications* 180, 2175-2196 (2009)
- [2] V. Kapil, M. Rossi et al.: *i-PI 2.0: A universal force engine for advanced molecular simulations*. *Computer Physics Communications* 236, 214-223 (2019)
- [3] Y. Litman, J. Richardson, T. Kumagai, and M. Rossi: *Elucidating the Nuclear Quantum Dynamics of Intramolecular Double Hydrogen Transfer in Porphycene*. *Journal of the American Chemical Society* 141, 2526-2534 (2019)
- [4] N. Raimbault, V. Athavale and M. Rossi: *Anharmonic effects in the low-frequency vibrational modes of aspirin and paracetamol crystals*, *Physical Review Materials* 3, 053605 (2019)
- [5] H. Wang, S.V. Levchenko, T. Schultz, N. Koch, M. Scheffler, and M. Rossi: *Modulation of the Work Function by the Atomic Structure of Strong Organic Electron Acceptors on H-Si(111)*. *Advanced Electronic Materials* 5, 1800891 (2018)

### **“Use the force!” reduced variance estimators for radial distribution functions, generic 3D densities and (local) transport coefficients**

**Benjamin Rotenberg**

Sorbonne University, Paris - France

**Room Lyon, 10:30**

**Tuesday 10**

A major goal of atomistic or coarse-grained simulations is to estimate physical properties from microscopic configurations. Given the computational cost to generate the latter, we want to make the most of our simulation data. Can we improve our estimates of a property for a given sample of configurations? This is the case for some key observables routinely computed to analyze the microscopic structure, namely radial distribution functions and local densities. The standard approach is to sample atomic positions using histograms. Such simple estimators are plagued with a diverging variance as the grid spacing goes to zero. I will illustrate how to obtain reduced variance estimators for radial distribution functions and generic (number, charge or polarization) three-dimensional densities, using the force on the atoms in addition to their position. Since the force is computed anyway in molecular dynamics simulations to generate the trajectory, not using it is a waste of valuable information. The benefit of this approach is even larger when the computational cost of producing the trajectory is high, in particular with ab initio MD. This presentation will introduce the basics [1] as well as recent [2] and on-going extensions of this idea of using the force to improve estimators.

[1] D. Borgis, R. Assaraf, B. Rotenberg, R. Vuilleumier, *Mol. Phys.* **111**, 3486 (2013).

[2] S.W. Coles, D. Borgis, R. Vuilleumier B. Rotenberg, *Computing three-dimensional densities from force densities improves statistical efficiency*. Submitted [<http://arxiv.org/abs/1905.11696>].

### **Quantum-cavity and floquet-engineered new states of matter from a QEDFT perspective**

**Angel Rubio**

Max Planck Institute for the Structure and Dynamics of Matter, Hamburg - Germany

**Room Orsay, 15:35**

**Tuesday 10**

Computer simulations that predict the light-induced change in the physical and chemical properties of complex systems usually ignore the quantum nature of light. Recent experiments at the interface between materials science and quantum optics have uncovered situations where both the molecular system and the photon field have to be treated in detail. In this talk, we show how the effects of quantum-photons can be properly included in the newly developed quantum electrodynamics density-functional formalism (QEDFT). We provide an overview of how well-established concepts in the fields of quantum chemistry and material sciences have to be adapted when the quantum nature of light becomes important. We identify fundamental changes in Born-Oppenheimer surfaces, conical intersections, spectroscopic quantities, and quantum control efficiency. We also show how periodic driving of many-body systems allow to design Floquet states of matter with tunable electronic properties on ultrafast time scales (and cavity induced-topology). This work paves the road for the development of two new fields, namely QED-materials and QED-chemistry.

*In collaboration with: H. Appel, M. Ruggenthaler, H. Hübener, U. de Giovannini, M. Sentef, J. Flick, C. Schafer, V. Rokaj, D. Welakuh, S. Sato, N. Park.*



## **Data-driven discovery of novel two-dimensional materials**

**Kristian Sommer Thygesen**

Technical University of Denmark, Kgs. Lyngby - Denmark

**Room Orsay, 11:00**

**Tuesday 10**

Atomically thin two-dimensional (2D) materials represent a rapidly moving frontier of condensed matter physics. The reduced phase space available and the low dielectric screening in these extremely thin materials lead to enhanced many-body effects and unique physics, which can be further tuned by external probes or by stacking the 2D materials into van der Waals heterostructures. In the first part, I will describe how classical electrostatic models can be combined with ab-initio quantum mechanics to describe novel types of excitonic and plasmonic states in 2D heterostructures. I will further illustrate various new concepts for band structure engineering in 2D heterostructures, which build on the detailed control of the internal electric fields and dielectric screening achievable in atomically thin materials[1]. In the second part of the talk I will present our recent efforts to establish a comprehensive computational database of 2D materials using an automatic high-throughput framework based on the Atomic Simulation Environment (ASE) (<http://c2db.fysik.dtu.dk>)[2] and demonstrate how it can be used to identify new 2D materials with interesting physical properties such as ferromagnetism and non-trivial band structure topologies[3].

[1] *Calculating excitons, plasmons, and quasiparticles in 2D materials and van der Waals heterostructures*, K. S. Thygesen, *2D Materials* ; 4 , 022004 (2017).

[2] *The Computational 2D Materials Database: High-throughput modeling and discovery of atomically thin crystals*, S. Hastrup ; et al. *2D Materials* 5 , 042002 (2018).

[3] *Discovering two-dimensional topological insulators from high-throughput computations*, T. Olsen, et al. *Phys. Rev. Mat.* ; 3 , 024005 (2019).

## **In silico synthesis of microgels: structure, elasticity and effective interactions in bulk and at liquid-liquid interfaces**

**Emanuela Zaccarelli**

University of Rome la Sapienza, Rome - Italy

**Room Blaricum, 15:05**

**Tuesday 10**

Microgels are soft particles individually made by cross-linked polymer networks which are nowadays widely used as a colloidal model system because of their swelling properties and their responsivity to external control parameters such temperature or pH. While extensively used as model systems in experimental, their numerical investigation lagged behind due to the inherently complex and multiscale nature of the particles. In this talk I will illustrate the protocol that we recently developed to synthesize microgels in-silico[1,2], providing a realistic description of the particles, in particular their characteristic inhomogeneous core-corona structure and their swelling behavior. I will also report on the calculation of their elastic properties and effective interactions in bulk [3] and at liquid-liquid interfaces[4]. The numerical results will be compared to available experiments. Our work aims to establish a clear link between the microscopic network properties and the resulting microgel-microgel interactions, paving the way for a deeper understanding of the behaviour of microgel suspensions.

[1] N. Gnan et al *Macromolecules* (2017).

[2] A. Ninarello et al *arxiv:1901.11495* (2019).

[3] L. Rovigatti et al *Macromolecules* (2019).

[4] F. Camerin et al *ACS Nano* (2019); in preparation.

## **Contributed talks**

### ***Ion-induced long-range orientational correlations in highly diluted electrolytes: what do second harmonic scattering experiments measure?***

**Daniel Borgis**<sup>1</sup>, Luc Belloni<sup>2</sup>, Maximilien Levesque<sup>3</sup>

<sup>1</sup>CNRS, Ecole Normale Supérieure de Paris and Maison de la Simulation, Saclay - France

<sup>2</sup>CEA, Saclay - France

<sup>3</sup>CNRS, Ecole Normale Supérieure de Paris - France

**Room Lyon, 12:20**

**Tuesday 10**

Recent second harmonic scattering measurements for highly diluted aqueous electrolytes show a 30% enhancement of the signal for salt concentrations as low as 50  $\mu$ molar (one ion pair for one million water molecules!) [1]. Following those experiments, ion-induced long-range orientational order in low concentration aqueous electrolytes has been evidenced using large-scale molecular dynamics simulations and looking at the averaged quantity  $\langle \cos \phi(r) \rangle$ ,  $\phi(r)$  the angle between the dipoles of two water molecules at distance  $r$  [2]. To decipher the observed phenomena, we have derived a theoretical expression of the second harmonic scattering signal in diluted electrolytes compared with bulk water [3,4]; the electrolyte structure is described through molecular integral equations theory which is exact at long distances [4]. We show that the enhancement of the signal with respect to pure water observed in the micromolar range is a mere manifestation of the Debye screening that makes the infinite-range dipole-dipole solvent correlations in  $1/r^3$  disappear as soon as the ionic concentration becomes finite. In  $q$ -space, this translates into a correlation function having a well known singular behaviour around  $q=0$ , which drives the observed ionic effects. We show that the signal is independent of the ion-induced long-range behaviour of the function  $\langle \cos \phi(r) \rangle$  that had been previously invoked. We find also that the enhancement depends on the experimental geometry and occurs only for in-plane polarisation detection, as observed experimentally. On the contrary, the measured isotope effect between light and heavy water cannot be fully explained.

[1] Chen et al. *Sci. Adv.* **2**, e1501891 (2016).

[2] Pluharova et al., *J. Phys. Chem. Lett.* **8**, 2031 (2017).

[3] Belloni et al., *J. Phys. Chem. Lett.* **9**, 1985 (2018).

[4] Borgis et al., *J. Phys. Chem. Lett.* **9**, 3698 (2018).

### ***From density functional theory to strength and ductility of complex metal alloys***

**William A. Curtin**

Institute of Mechanical Engineering, Swiss Federal Institute of Technology (EPFL), Lausanne - Switzerland

**Room Lausanne, 16:35**

**Tuesday 10**

When solute atoms are added to elemental metals to create engineering alloys, they have a range of effects on the behavior of the dislocations that are the carriers of plasticity in metals. These effects then determine the alloy strength and ductility, which are crucial engineering-scale metrics for applications. Direct first-principles simulations of the behaviors of interest are completely infeasible due to size and time limits. Even methods using semi-empirical interatomic potentials are not very useful, especially due to the lack of quantitative potentials for alloys.

Quantitative progress is thus made through the development of mechanistic theories of the key phenomena controlling strength or ductility combined with the application of first-principles density function theory (DFT) to compute the inputs to these theories with chemically-specific accuracy. Here, we briefly discuss recent successes in the prediction of (i) ductility in dilute Mg alloys and (ii) yield stresses in fcc High Entropy Alloys. The theories combined with DFT inputs then enable a priori predictions of new alloy compositions for enhanced performance across a very broad compositional space. Several examples are presented and they show that the long-standing promise of computationally-guided alloy design for mechanical performance is becoming a reality.

### **Fluctuating hydrodynamics and debye-hückel-onsager theory for electrolytes**

**Aleksandar Donev**<sup>1</sup>, Alejandro L. Garcia<sup>2</sup>, Jean-Philippe Péraud<sup>3</sup>, Andy Nonaka<sup>3</sup>, John B. Bell<sup>3</sup>

<sup>1</sup>Courant Institute, New York University, New York - USA

<sup>2</sup>San Jose State University, San Jose - USA

<sup>3</sup>Lawrence Berkeley National Laboratory, Berkeley - USA

**Room Lyon, 12:00**

**Tuesday 10**

We apply fluctuating hydrodynamics to strong electrolyte mixtures to compute the concentration corrections for chemical potential, diffusivity, and conductivity. We show these corrections to be in agreement with the limiting laws of Debye, Hückel, and Onsager. We compute explicit corrections for a symmetric ternary mixture and find that the co-ion Maxwell-Stefan diffusion coefficients can be negative, in agreement with experimental findings.

### **Symmetry-inspired mesoscopic models for studying partially-ordered mesophases of semiconducting polymers**

**Kostas Daoulas**, Cristina Greco, Anton Melnyk, Kurt Kremer, Denis Andrienko,  
Max Planck Institute for Polymer Research, Mainz - Germany

**Room Lausanne, 16:55**

**Tuesday 10**

The rich phase behavior of soluble semiconducting polymers makes them interesting for basic polymer physics, because it allows one to access partially-ordered states – microstructures located on the order-disorder scale between (poly)crystalline and amorphous morphologies. Studying microstructures with varying degree of order can facilitate, for example, understanding various phenomena in polymer crystallization. Technologically, partially-ordered phases are interesting because they are relevant for new-generation materials with improved electronic properties [1].

We develop a computationally-efficient simulation strategy [2,3] for investigating the molecular organization in partially-ordered mesophases. The molecular architecture is described by chains of coarse-grained monomers. Each monomer represents (at least) one atomistic repeat unit, i.e. it “encapsulates” the cyclic moieties of the backbone and side chains. Soft anisotropic non-bonded potentials capturing co-planarity in chain orientation and stacking are a novel methodological feature of our approach [3]. The potentials are developed using generic symmetry arguments. The model provides a framework for efficient Monte Carlo simulations. We demonstrate that the method describes a broad spectrum of morphologies, including amorphous, nematic, and lamellar-like, smectic, mesophases. We extensively discuss the molecular organization of these morphologies, focusing on smectic mesophases. The lamellar order is characterized through various quantifiers including scattering patterns, analogous to experimental GIWAXS. We demonstrate that our smectic mesophases match morphologies that have been experimentally observed in P3HT [4]. We investigate the influence of conformational properties and mesoscale organization of polymer on charge-carrier mobility introducing a qualitative charge-transport model.

[1] R. Noriega, J. Rivnay, K. Vandewal, F.P.V. Koch, N. Stingelin, P. Smith, M.F. Toney, and A. Salleo, *Nat. Mater.* **12**, 1038 (2013).

[2] P. Gemünden, C. Poelking, K. Kremer, D. Andrienko, and K.C. Daoulas, *Macromolecules* **46**, 5762 (2013).

[3] C. Greco, A. Melnyk, K. Kremer, D. Andrienko, and K.C. Daoulas, *Macromolecules* **52**, 968 (2019).

[4] Z. Wu, A. Petzold, T. Henze, T. Thurn-Albrecht, R.H. Lohwasser, M. Sommer, and M. Thelakkat, *Macromolecules* **43**, 4646 (2003).

### **Oxygen-vacancy ordering and dynamics at the reduced CeO<sub>2</sub>(111) surface and the entanglement with polaron hopping**

**M. V. Ganduglia-Pirovano**<sup>1</sup>, G. E. Murgida<sup>2</sup>, A. M. Llois<sup>2</sup>, V. Ferrari<sup>3</sup>, D. Zhang<sup>4</sup>, Z-K. Han<sup>5</sup>, Y. Gao<sup>5</sup>

**Room Orsay, 12:20**

**Tuesday 10**

<sup>1</sup>Centro Atómico Constituyentes, CNEA-CONICET, San Martín, Buenos Aires - Argentina

<sup>2</sup>Instituto de Catálisis y Petroleoquímica, CSIC, Madrid - Spain

<sup>3</sup>Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai - China

<sup>4</sup>Fritz Haber Institute, Max Planck Society, Berlin - Germany

<sup>5</sup>Shanghai Advanced Research Institute, Chinese Academy of Sciences, Shanghai - China

The ability of ceria ( $\text{CeO}_2$ ) to store, release, and transport oxygen is crucial to its functionality in applications in catalysis, fuel cells, sensors, and recently in biology. Deep understanding of the structure and dynamics of oxygen vacancies at ceria surfaces is essential for the rational design of improved systems in such applications. For the ceria (111) surface, whether oxygen vacancies prefer the subsurface or the surface and if surface oxygen vacancies attract or repel as well as whether oxygen migration and polaron ( $\text{Ce}^{3+}$ ) hopping are entangled, are still heavily debated. Also, a number of ordered phases have been observed upon reduction, but their structures have remained elusive. Here, supported by experimental and theoretical results, obtained employing density functional theory in combination with statistical thermodynamics, Monte Carlo simulations, and ab-initio molecular dynamics, the current understanding of the structure of the reduced ceria (111) surface will be discussed [1-7]. We predict the preference of polarons for next-nearest cationic sites to the vacancies [1-3], and that at low temperature and low vacancy concentrations, vacancies prefer subsurface sites with a local (2x2) ordering, whereas mostly linear surface vacancy clusters do form with increased temperature and degree of reduction [3,7]. Moreover, we explain the nature of the observed reduced ceria (111) surface reconstructions [4-6]. Finally, in the 300-900 K range, we identify different dynamic regimes in the migration of oxygen vacancies and in the hopping of polarons and show that their dynamic behaviors are entangled [8].

[1] M.V. Ganduglia-Pirovano, J.L.F. Da Silva, J. Sauer, *Phys. Rev. Lett.* **102**, 026101 (2009).

[2] J.F. Jerratsch, X. Shao, N. Nilius, H.J. Freund, C. Popa, M.V. Ganduglia-Pirovano, A.M. Burow, J. Sauer, *Phys. Rev. Lett.* **106** 246801 (2011).

[3] G.E. Murgida, M.V. Ganduglia-Pirovano, *Phys. Rev. Lett.* **110**, 246101 (2013).

[4] G.E. Murgida, V. Ferrari, A.M. Llois, M.V. Ganduglia-Pirovano, *Phys. Rev. B* **90**, 115120 (2014).

[5] R. Olbrich, G.E. Murgida, V. Ferrari, C. Barth, A.M. Llois, M. Reichling, M.; M.V. Ganduglia-Pirovano, *J. Phys. Chem. C*, **121**, 6844 (2017).

[6] Murgida, G. E.; Ferrari, V.; Llois, A. M.; M.V. Ganduglia-Pirovano, M. V. *Phys. Rev. Materials* **2**, 083609 (2018).

[7] Han, Z.-K.; Yang, Y.-Z.; Zhu, B.; Ganduglia-Pirovano, M. V.; Gao, Y. *Phys. Rev. Materials* **2**, 035802 (2018).

[8] Zhang, D.; Han, Z.-K.; Murgida, G. E.; M.V. Ganduglia-Pirovano, M. V.; Gao, Y. *Phys. Rev. Lett.* **122**, 019601 (2019).

**OH!**

**Kersti Hermansson**

Uppsala University, Uppsala - Sweden

**Room Orsay, 11:30**

**Tuesday 10**

Hydroxylation & hydration of solid surfaces deserve special attention in materials science as these phenomena can greatly modify the surface properties – in a detrimental or beneficial fashion. I will present some efforts to use physics-based and data-driven models to explore the structure of metal oxide/water interfaces. The former methods provide insight. The latter open up for the harvesting of massive amounts of interface data which can generate properties and spectra with outstanding statistics and resolution. A few examples are given in Refs [1]-[3]. In the course of this work we were also able to discover a descriptor which helps to distinguish between OH vibrational frequency signals from intact and dissociated water molecules; these generally fall in very much the same frequency regions which makes them difficult to distinguish.

[1] G. G. Kebede, P. D. Mitev, K. Hermansson, *To be submitted* (2019).

[2] A. Röckert, P.D. Mitev, J. Kullgren, K. Hermansson. *To be submitted* (2019).

[3] V. Quaranta, M. Hellström, J. Behler, J. Kullgren, P.D. Mitev, and K.Hermansson, *J. Chem. Phys.* **148**, 241720 (2018). (*Editor's choice* 2018).

## **Molecules at the electrochemical interface: understanding experiments with simulations**

Marcella Iannuzzi, Gabriele Tocci, Tiziana Musso

University of Zürich, Zürich - Switzerland

Room Orsay, 16:55

Tuesday 10

The fundamental understanding of interfaces at the atomic level would greatly help in predicting the stability and reactivity of materials and eventually would lead to the rational tuning of the different components for example for heterogeneous catalysts, nano-electronics, photocatalysis. Most of the relevant processes in an electrochemical cell occur at solid/liquid interfaces. Unfortunately, the direct observation of these processes and the characterisation of the interfaces under operating conditions are still challenging for both experimental and modeling approaches. However, by developing efficiently scaling computational strategies, modelling of materials properties and processes from first principles is becoming sufficiently accurate as to facilitate the design and testing of new systems in silico. We present a few examples where computational materials science turns out to be valuable and necessary for developing novel functional materials. In order to shed some light on selected functionalised electrochemical interfaces, we employ electronic structure calculations based on density functional theory, combined with the simulation of spectroscopic properties and with ab initio molecular dynamics (AIMD)[1]. In particular, we present our studies on the functionalisation of supported hexagonal boron nitride (h-BN)[2], which modifies the interaction of adsorbing species on the metal, their reactivity and spectroscopic signature. We identify by orbital imaging the adsorption of reactive species - monodisperse triangular  $(\text{WO}_3)_3$  clusters - which experimentally is achieved at room temperature and in the presence of a liquid electrolyte [3]. Here, h-BN acts as atomically thin insulator, decoupling the cluster from the metal but allowing its reduction to  $[\text{W}_3\text{O}_9]^{2-}$  by charge transfer into the W-d orbitals. We also show how the nanotexture of h-BN (corrugated vs flat by H-intercalation) affects these properties and how this can be related to the exfoliation of the functionalised two dimensional membrane. It has been shown that such a membrane can be employed for blue energy harvesting like osmotic power generation or ion separation[4]. Hence, AIMD simulations are carried out to understand the fluid structure and the dynamics at the interface with such 2D materials, with aiming at finding the relation between the electronic structure and the osmotic transport.

[1] J. Hutter, J. M. Iannuzzi, M., F. Schiffmann, F., J. VandeVondele, J. Wiley Interdisciplinary Reviews: Computational Molecular Science **4**, 15 (2014).

[2] S.F.L. Mertens, A. Hemmi, S. Muff, O. Gröning, S. DeFeyter, J. Osterwalder, T. Greber, Nature **534**, 676 (2016).

[3] M. Muellner, J. Balajka, M. Schmid, U. Diebold, S.F.L. Mertens, J. Phys. Chem. C **121**, 19743 (2017).

[4] H. Cun, A. Hemmi, E. Miniussi, C. Bernard, B. Probst, K. Liu, K., et al., Nano Letters **18**, 1205 (2018).

## **Vibrational excitations and elastic response at the nanoscale: from disordered solids to nanostructures**

Stefano Mossa<sup>1</sup>, Hideyuki Mizuno<sup>2</sup>, Jean-Louis Barrat<sup>3</sup>

<sup>1</sup>CEA Grenoble - Interdisciplinary Research Institute of Grenoble (IRIG), Grenoble - France

<sup>2</sup>The University of Tokyo, Tokyo - Japan

<sup>3</sup>University Grenoble Alpes, Saint-Martin-d'Hères - France

Room Orsay, 16:35

Tuesday 10

In a recent series of papers [1-5] we have described extensive numerical work focusing on the concept of elastic heterogeneity in disordered solids. The fundamental idea is quite simple: glasses and even (complex) crystals with defects show inhomogeneous mechanical response at the nanoscale, which does not conform to the macroscopic limit predictions. This property can be demonstrated to strongly influence vibrational and thermal properties of the materials, and could ultimately lie at the bottom of puzzling anomalous features like boson peak, Raleigh-like strong scattering, or temperature dependence of thermal conductivity. We have investigated the above issues systematically and in great details by classical Molecular Dynamics simulation. On one side, we have employed different toy models including monodisperse Lennard Jones nanoparticles, and soft spheres with particles size disorder. On the other, we have considered multiple phases with variable amount of disorder, ranging from glassy configurations, to crystals with defects and superlattices. In all cases, we have monitored independently mechanical response, collective excitations, and heat transfer features, establishing precise correlations among the heterogeneous local mechanical response, the nature of the vibrational states, and the variation of thermal conductivity. In the talk I will discuss some of our findings and put them in perspective, also with reference

to advanced spectroscopy measurements with X-Rays. I will also show how information of this nature can be exploited to engineer interfaces, devising metamaterials with tailored functions.

1. H. Mizuno and S. Mossa, *Sound broadening in glasses: interaction with anharmonicities and elastic heterogeneities*, submitted (2019).
2. H. Mizuno, S. Mossa, and J.-L. Barrat, *Phys. Rev. B* **94**, 144303 (2016) [10.1103/PhysRevB.94.144303].
3. H. Mizuno, S. Mossa, and J.-L. Barrat, *Sci. Rep.* **5**, 14116 (2015) [10.1038/srep14116].
4. H. Mizuno, S. Mossa, and J.-L. Barrat, *Proc. Natl. Acad. Sci.* **111**, 11949 (2014) [10.1073/pnas.1409490111].
5. H. Mizuno, S. Mossa, and J.-L. Barrat, *EPL* **104**, 56001 (2013) [10.1209/0295-5075/104/56001].

### **Simultaneous sampling of multiple transition channels using adaptive paths of collective variables**

**Alberto Pérez de Alba Ortíz, Bernd Ensing**

**Room Lausanne, 12:20**

**Tuesday 10**

We present a novel molecular simulation framework to perform simultaneous free energy calculations along multiple repulsive and adaptive paths of collective variables, and thus find the mechanisms and probabilities of competing molecular transition pathways. The scheme relies on a multiple-path multiple-walker implementation of path-metadynamics (multiPMD), in which the parallel walkers are grouped to sample simultaneously different mechanistic pathways. To discourage that the different groups localize into the same, most likely, pathway, each group contains a special, non-metadynamics, walker that repels those from the other groups. Interestingly, these non-metadynamics walkers can also pinpoint known distinctive landmarks, while keeping the rest of the path flexible. We illustrate the multiPMD framework for two C7eq-to-C7ax paths in alanine dipeptide and for six PPI-to-PPII paths in tetrameric polyproline. We also propose a variation for the treatment for cyclic or periodic paths, which we apply to a DNA base-pairing transition with alternative pathways. MultiPMD adds to the performance of path-based biasing methods by enabling a parallel search for routes beyond the minimum free energy path or the most-likely transition path, hence broadening our perspective on complex transition networks with multiple collective variables. The scheme is highly flexible regarding the number of paths and repellers, the nature of the repulsive potential, and the choice of the biasing method along the paths.

[1] G. Díaz Leines, and B. Ensing, *Phys. Rev. Lett.* **109**, 020601 (2012).

[2] A. Pérez de Alba Ortíz, A. Tiwari, R. C. Puthenkalathil, and B. Ensing, *J. Chem. Phys.* **149**, 072320 (2018).

[3] A. Pérez de Alba Ortíz, J. Vreede, and B. Ensing, in *Biomolecular Simulations: Methods and Protocols*, edited by M. Bonomi, and C. Camilloni (Springer, 2019).

[4] A. Pérez de Alba Ortíz, and B. Ensing, (In prep.).

### **Phoretic colloids and micromachines**

**Marisol Ripoll**

Institute Complex Systems, Forschungszentrum Jülich, Jülich - Germany

**Room Blaricum, 12:00**

**Tuesday 10**

Thermophoresis refers to the directed motion of colloidal particles in a temperature gradient. Together with the colloid motion, the temperature gradient also induces a thermoosmotic flow of the surrounding solvent. This flow can eventually translate into the formation of thermophoretic crystals, or be used to generate diverse flow patterns in microfluidic environments. The thermophoretic effect can also be exploited to build micromachines, which we investigate by means of a mesoscopic simulation technique known as multiparticle collision dynamics simulations (MPC). Asymmetric microgears locally heated in a cooled surrounding solvent can be shown to rotate spontaneously and unidirectionally. Microscale turbines rotating in the presence of external fields can be constructed by assembling anisotropic blades in a chiral manner based in the so-called anisotropic thermophoretic effect, characteristic of elongated objects. Self-propelled motion due to thermophoresis can be induced for particles with asymmetric properties such as Janus or dimer colloids, which display very interesting single and collective hydrodynamic behavior such as clustering or swarming. Interestingly, most of these concepts are also valid in the case of diffusiophoresis which relies on the intrinsic existence of concentration gradients, as those produced in the presence of catalytic surfaces.

***From chromosome territories to ring polymers: physical properties of untangled polymers melts***

Angelo Rosa<sup>1</sup>, Ralf Everaers<sup>2</sup>

<sup>1</sup>Scuola Internazionale Superiore di Studi Avanzati - SISSA, Trieste - Italy

<sup>2</sup>Ecole Normale Supérieure de Lyon, Lyon - France

**Room Blaricum, 12:20**

**Tuesday 10**

In this talk, I will review my work on the physical modeling of eukaryotic chromosomes. In particular, I will present results of detailed molecular dynamics computer simulations of a minimalistic coarse-grained polymer model which is able to reproduce with great accuracy the large-scale features of chromosomes, like their confinement to specific regions of the nucleus (territories) and the formation of chromatin contacts. The talk will be concluded by a discussion focusing on the conceptual connection between nuclear chromosome organization and the physics of untangled ring polymers in concentrated solutions.

***Shape, height fluctuations and resisting force of a membrane deformed by an actin bundle***

Jean Paul Ryckaert

Université Libre de Bruxelles, Brussels - Belgium

**Room Blaricum, 16:55**

**Tuesday 10**

We consider a triangulated membrane, with surface tension and bending energy parametrized to represent the physiologic double layer of lipids of the cell membrane, strongly deformed by a single actin rigid filament or by a bundle of parallel filaments, hence forming a local finger-like structure where the membrane envelops the bundle, a structure reminiscent of filopodia found in vivo. The membrane coarse grain degrees of freedom are sampled in the presence an external field due to fixed filaments pointing normally to the unperturbed membrane surface: we use a fixed tension, fixed temperature and fixed projected area grand canonical Monte-Carlo method proposed in [1]. We determine for various filament tip relative heights configurations, the average shape of the deformed membrane and its equilibrium fluctuations. For the adopted hard-core interaction model, the tip-membrane gap size distribution is the central observable as it yields both the average force exerted by the membrane on the tip and the probability for a filament to accept a polymerization step against the equilibrated deformed membrane. These data are exploited to predict the bundle growth/shrinking dynamics of a well developed filopodium for an ideal brownian ratchet model where a large time scale separation between the chemical steps and the much faster local membrane relaxation is assumed.

[1] J. Weichsel and Ph. Geissler, *PLOS Comp. Biol.* **12**, e1004982 (2016).

***Prebiotic chemistry and origins of life studies through ab initio calculations***

Antonino Marco Saitta, Fabio Pietrucci

Sorbonne University, Paris - France

**Room Lyon, 16:35**

**Tuesday 10**

Computational approaches are nowadays a full, self-standing branch of chemistry, both for their quantum-based (“ab initio”) accuracy, and for its multiscale extent. In prebiotic chemistry, however, due to the intrinsic complexity of the chemical problems, ab initio atomistic simulations have so far had a limited impact, with the exception of a few relevant studies [1], including the elucidation of the chemical interactions between biomolecules with surfaces, such as ice and minerals, or the simulation of the effect of the pressure/temperature shock waves induced by meteorite impacts in the early Earth. Surprisingly, even the celebrated Miller experiments, which historically reported on the spontaneous formation of amino-acids from a mixture of simple molecules reacting under an electric discharge, had never been studied at the quantum atomistic level.

Here we set the general problem of chemical networks within new topology-based concepts, which allows a very efficient definition of reaction coordinates even in the complex chemical environments typical of likely prebiotic scenarii. We thus report on the first ab initio computer simulations, based on quantum physics and a fully atomistic approach, of Miller-like experiments in the condensed phase. Our study [2]

shows that glycine spontaneously form from mixtures of simple molecules once an electric field is switched on. We identify formic acid and formamide [3] as key intermediate products of the early steps of the Miller reactions, and the crucible of formation of complex biological molecules, as confirmed by our recent experimental and theoretical study on high-energy chemistry of formamide [4]. From a broader chemical perspective, we show that formamide plays the role of hub of a complex reaction network in both the gas and the condensed phase [5]. We are now going on a larger scale, studying the atomistic mechanisms of RNA nucleotides synthesis [6], meteoritic amino acids [7] and sugars [8] in fully realistic prebiotic solution environments. All these results pave the way to novel computational approaches in the research of the chemical origins of life.

[1] A. Pérez-Villa, F. Pietrucci, A.M. Saitta *Physics of Life: Reviews* doi.org/10.1016/j.plrev.2018.09.004 (2019).

[2] A.M. Saitta and F. Saija *Proceedings of the National Academy of Sciences USA* **111**, 13768 (2014) .

[3] A.M. Saitta, F. Saija, F. Pietrucci, and F. Guyot, *Proceedings of the National Academy of Sciences USA* **112**, E343 (2015).

[4] M. Ferus et al. *Proceedings of the National Academy of Sciences USA* **114**, 4306 (2017).

[5] F. Pietrucci, and A.M. Saitta, *Proceedings of the National Academy of Sciences USA* **112**, 15030 (2015).

[6] A. Perez-Villa et al., *J. Phys. Chem. Lett.* **9**, 4981 (2018).

[7] F. Pietrucci et al., *ACS Earth Space Chem* **2**, 588 (2018).

[8] G. Cassone et al., *Chem Comm* **54**, 3211 (2018).

### **Biosimulation, 25 years of**

**Marie Christine Sawley**

Intel Semiconductor AG, Zürich - Switzerland

**Room Lausanne, 12:00**

**Tuesday 10**

Powerful computers have always been the close companion of Material and molecular sciences. The evolution of the underlying computing technology has fueled innovation and creativity in many scientific domains, but perhaps the most remarkable leap forward of the last 20 years took place in biosimulation and biological data analysis. Recent increased capacities deriving from the combination of HPC and Data Analytics scalable technology is pushing further the boundaries of discoveries and frontiers. This talk will show with concrete examples how extreme computing technology has enabled progress in biosimulation, while being inspired by the rich portfolio of life science through co design activities; it will also open the perspective on how powerful and innovative technologies such as quantum computing and neuromorphic processors may enable the expansion during the next decades. (*The author is a computational physicist who has been working throughout her career at the crosspoint of leading edge material science and extreme computing technologies*).

### **Challenges, advances and perspectives of protein aggregation simulations**

**Birgit Strodel<sup>1</sup>**, Martin Carballo-Pacheco<sup>2</sup>, Bogdan Barz<sup>1</sup>, Ushnish Sengupta<sup>3</sup>

<sup>1</sup>Forschungszentrum Jülich, Jülich - Germany

<sup>2</sup>University of Edinburgh, Edinburgh - United Kingdom

<sup>3</sup>Department of Engineering, University of Cambridge, Cambridge - United Kingdom

**Room Lyon, 16:55**

**Tuesday 10**

The aim of our work is to understand the physicochemical principles that govern the highly complex process of protein aggregation. All-atom molecular dynamics (MD) simulations of protein aggregation in explicit solvent have been performed for more than 15 years, revealing valuable information about this phenomenon. However, these simulations are challenged by three main problems: (1) The accuracy of current all-atom force fields in modeling protein aggregation is insufficient [1]. We presently work on elucidating why the force fields fail to reproduce the aggregation kinetics, so that in a next step we can resolve these problems by adapting the force fields. (2) The second problem is that MD simulations of protein aggregation are generally performed at protein concentrations orders of magnitude higher than the comparable in vitro and in vivo situations, limiting structural rearrangements between aggregate growth events [2]. In order to overcome this limitation we developed Markov state models of protein aggregation and showed that at least tens of microseconds of MD data is needed to sample the phase space of the individual aggregation states and their mutual transformation [3]. (3) The third problem is the well-known length- and time-scale problem of molecular simulations, limiting their comparability to



experiments that usually report on nm-to- $\mu\text{m}$  long aggregates forming on the time scale of minutes and beyond [4]. This limitation can only be solved by designing experiments that also elucidate the early aggregation events, and by developing multiscale simulation approaches that will allow to extend the simulated length- and time-scales. To this end, a multiscale simulation method with the aggregating proteins being presented at the atomistic level and their environment being described by a coarse-grained force field is under development in the Strodel group. In the long run, this multiscale approach will facilitate protein aggregation under in vivo conditions to be simulated.

[1] M. Carballo-Pacheco, A. E. Ismail, B. Strodel, *J. Chem. Theory Comput.* **14**, 6063 (2018).

[2] M. Carballo-Pacheco and B. Strodel, *J. Phys. Chem. B* **120**, 2991 (2016).

[3] U. Sengupta, M. Carballo-Pacheco, B. Strodel, *J. Chem. Phys.* **150**, 115101 (2019).

[4] B. Barz, Q. Liao, B. Strodel, *J. Am. Chem. Soc.* **140**, 319 (2018.)

### **First-principles prediction of vibronic spectra, internal conversion, and wavelength-dependent product quantum yields**

**Enrico Tapavicza**

California State University Long Beach, Long Beach - USA

**Room Orsay, 12:00**

**Tuesday 10**

I will present a time-dependent generating function approach to compute vibronic absorption and emission spectra [1]. The method takes into structural changes between ground and excited state geometries and fully incorporates Duschinsky mode mixing effects. An extension to this approach is presented that allows to compute individual vibronic transitions from singly-excited vibrational states [2]. This allows to simulate single collision free vibronic level (SVL) fluorescence spectra and temperature effects in electronic spectra. Besides, this algorithm can also be used to compute radiationless decay of excited states. Methods are applied to polyaromatic hydrocarbons and organic dyes. As second topic, I will focus on the conformational dependence in electronic spectra [3,4] and the prediction of wavelength-dependent product quantum yields of conformationally controlled photochemical reactions via non-adiabatic molecular dynamics [5,6]. Applications include organic aerosols [4], photoswitches [6], and vitamin D photochemistry [3,5].

[1] E. Tapavicza, F. Furche, D. Sundholm, *J. Chem. Theory Comput.* **12**, 5058 (2016).

[2] E. Tapavicza, L. Li, *in preparation*.

[3] C. Cisneros, T. Thompson, N. Baluyot, A. C. Smith, E. Tapavicza *Phys. Chem. Chem. Phys.* **19**, 5763 (2017).

[4] D. O. De Haan, E. Tapavicza, M. Riva, T. Cui, J. D. Surratt, A. C. Smith, M. Jordan, S. Nilakantan, M. Almodovar, T. N. Stewart, A. de Loera, A. C. De Haan, M. Cazaunau, A. Gratien, E. Pangui, J. Doussin, *Environ. Sci. Technol.* **52**, 4061 (2018).

[5] T. Thompson, E. Tapavicza, *J. Phys. Chem. Lett.* **9**, 4758 (2018).

[6] E. Tapavicza, T. Thompson, K. Redd, D. Kim, *Phys. Chem. Chem. Phys.* **20**, 24807 (2018).

### **Complex memory in soft materials**

**Francisco Vega Reyes**<sup>1</sup>, Antonio Lasanta<sup>2</sup>, Antonio Prados<sup>3</sup>, Andrés Santos<sup>4</sup>

<sup>1</sup>Departamento de Física, Universidad de Extremadura, Badajoz - Spain

<sup>2</sup>Universidad Carlos III, Madrid, Madrid - Spain

<sup>3</sup>Universidad de Sevilla, Sevilla - Spain

<sup>4</sup>Universidad de Extremadura, Badajoz - Spain

**Room Blaricum, 16:35**

**Tuesday 10**

Intuition from every day life tells us that the temperature  $T_s$  of a physical system keeps constant if the system is put in contact with a thermal reservoir at the same temperature  $T_s$ . However, under certain conditions, "memory" of the system history can be retained: according to research literature, for a system in such a situation the temperature may display a small departure (either increase or decrease) and then return back to the thermal reservoir value  $T_s$ . We show now that these memory effects can actually be quite more complex -comprising several extrema instead of a single maximum or minimum-, and display huge deviations from  $T_s$ . We explain the mechanism that leads to giant and complex thermal memory [1,2]. The mechanism is quite generic and can potentially be present in a variety of systems. As a case study, we describe the properties of large and complex thermal memory in a gas of inelastic and frictional

particles. The system is thermally excited by an external and homogeneous volume force. The analysis is performed via 3 independent methods: theoretical solution of the corresponding kinetic equation, molecular dynamics simulations and direct simulation of Monte Carlo of the kinetic equation.

[1] A. Lasanta, F. Vega Reyes, A. Prados and A. Santos, *Phys. Rev. Lett.* **119**, 148001 (2017).

[2] A. Lasanta, F. Vega Reyes, A. Prados and A. Santos, *New J. Phys.* **21**, 033042 (2019).

## Plenary speaker

### ***Next-generation first-principles based molecular dynamics: from biological systems to materials***

**Ursula Röthlisberger**

Swiss Federal Institute of Technology (EPFL), Lausanne - Switzerland

Room Lausanne, 09:00

Wednesday 11

Density Functional based molecular dynamics simulations and their quantum mechanical/molecular mechanical (QM/MM) extensions are powerful tools for the investigation of quantum mechanical phenomena in extended systems. However, the time scale that can be covered in such simulations is still limited even when they are used in combination with enhanced sampling techniques. To make first-principles based simulations more efficient; we have developed a new highly flexible interface for multiscale simulations in CPMD [1] together with novel schemes for the calculation of exact exchange within plane wave calculations [2]. Through the extensive use of multiple-time step techniques [3] and combinations with algorithms from artificial intelligence (AI), first-principles based molecular dynamics simulations can be sped up by several orders of magnitude. This combination of traditional computational chemistry methods with approaches from AI constitutes an exceptionally powerful recipe for next-generation first-principles based multiscale simulations. In this talk, some illustrative applications from biological systems to material science will be presented

[1] J. M. H. Olsen, V. Bolnykh, S. Meloni, E Ippoliti, M. P Bircher, P. Carloni and U. Rothlisberger, *MiMiC: A Novel Framework for Multiscale Modeling in Computational Chemistry*, *J. Chem. Theor Comp.* **15**, 3810-3823 (2019)

[2] M Bircher and U. Rothlisberger, *Exploiting Coordinate Scaling Relations to Accelerate Exact Exchange Calculations*, *J. Phys. Chem. Lett.* **9**, 3886-38890 (2018)

[3] E. Liberatore, R. Meli, and U. Rothlisberger, *A versatile multiple time step scheme for efficient Ab Initio molecular dynamics simulations*, *J. Chem. Theor. Comp.* **14**, 2834-2842 (2018)

## Invited speakers

### ***Understanding the dynamical bottlenecks in complex activated (bio)molecular processes***

**Peter Bolhuis**

University of Amsterdam, Amsterdam - Netherlands

Room Lausanne, 11:30

Wednesday 11

The investigation of rare but important events has always been a focal point in the field of molecular simulation, from the invention of umbrella sampling almost 50 years ago to the latest adaptive technique. Many of these methods rely on the definition of a collective variable that describes the progress of the transition sufficiently well to avoid sampling problems. In fact, the search for this reaction coordinate is at the heart of the rare event problem. Transition path sampling (TPS) avoids this problem by focusing on the pathways undergoing the activated event. Instead, the reaction coordinate can be extracted from the resulting path ensemble using variants of machine learning. Over the last twenty years, since its inception, many variants of TPS have been developed. Strangely enough, only recently it has become possible to efficiently perform large-scale TPS simulations on complex systems, such as clathrate nucleation and protein dissociation. Another strand of development is the transition interface sampling (TIS) methodology to compute rate constants, the application to multiple states, and adaptive path sampling. The development of the OpenPathSampling framework [1,2] has enabled easy and flexible use and implementation of novel path sampling algorithms on large systems. In this presentation, I will give a brief

overview of recent developments and discuss case studies on methane hydrate nucleation, DNA base pair rolling, and protein conformational changes.

[1] D.W.H. Swenson, J.-H. Prinz, F. Noe, J.D. Chodera, P.G. Bolhuis, *J. Chem. Theory Comput.* **15**, 813 (2019). DOI: 10.1021/acs.jctc.8b00626.

[2] D.W.H. Swenson, J.-H. Prinz, F. Noe, J.D. Chodera, P.G. Bolhuis *J. Chem. Theory Comput.*, 2019, **15**, 837 (2019). 856 DOI: 10.1021/acs.jctc.8b00627.

## **Exchange-correlation functionals for the band structure of solids: from bulk systems to interfaces and surfaces**

**Silvana Botti**

Friedrich-Schiller Universität Jena, Jena - Germany

**Room Orsay, 10:30**

**Wednesday 11**

I will first present the results of a recent benchmark study in which we evaluate the quality of exchange-correlation functionals for the determination of electronic band gaps of solids [1]. We compiled for this benchmark a dataset with information on the experimental structure and band gaps of 475 non-magnetic semiconductors. This dataset was used to test 12 functionals, ranging from standard local and semi-local functionals, passing through meta-generalized-gradient approximations, and some hybrids.

I will then consider two first-rate functionals for band structures, namely the screened hybrid functional HSE [2] and the modified Becke-Johnson (MBJ) [3] exchange-correlation potential, and I will propose how to modify them to calculate accurate band alignments and defect energy levels at interfaces.

We have in fact proposed a local hybrid functional for interfaces [4], extending the results of a previously developed density-dependent mixing for bulk crystals [5]. However, hybrid functional calculations can still be challenging for large supercells containing interfaces. The MBJ exchange-correlation potential would be in that case the ideal choice, but it can be used only for bulk crystals. We turned therefore the coefficient  $c$ , that multiplies the Becke-Russel exchange potential, into a local function of the position, along the lines of what we had already done for the mixing parameter of hybrid functionals. These local functionals yield results of comparable quality as GW at a significantly reduced cost. This is because we account properly for the position dependence of electronic screening, through a density estimator of the local di-electric function.

[1] P. Borlido, T. Aull, A.W. Huran, F. Tran, M.A.L. Marques, and S. Botti, *J. Chem. Theory Comput.* (2019). <https://doi.org/10.1021/acs.jctc.9b00322>.

[2] J. Heyd, G. E. Scuseria, and M. Ernzerhof, *J. Chem. Phys.* **118**, 8207 (2003); *J. Chem. Phys.* **124**, 219906 (2006).

[3] F. Tran and P. Blaha, *Phys. Rev. Lett* **102**, 226401 (2009).

[4] P. Borlido, M.A.L. Marques, and S. Botti, *J. Chem. Theory Comput.* **14**, 939-947 (2018).

[5] M.A.L. Marques, J. Vidal, M.J.T. Oliveira, L. Reining, and S. Botti, *Phys.Rev. B* **83**, 035119 (2011).

## **Looking inside a polaron**

**Feliciano Giustino**

University of Oxford, Oxford - United Kingdom

**Room Orsay, 11:00**

**Wednesday 11**

Polarons are among the most well-known quasiparticles in solid state physics, and are key to understanding fundamental concepts such as the electron mass enhancement in semiconductors and the formation of Cooper pairs in superconductors.

Polarons attracted considerable attention ever since the concept was formulated by Landau a century ago. During the past decades significant progress has been achieved in the numerical solution of model Hamiltonians describing polarons, however first-principles calculations are still scarce. This lag is probably due to some practical as well as fundamental challenges, namely that explicit DFT calculations of polarons require very large supercells; that polaron self-trapping might be missed due to the DFT delocalization problem; and that the polaron binding energies can be very sensitive to the choice of the exchange and correlation functional.

In this talk I will describe a new approach to the polaron problem that aims at overcoming these limitations [1,2]. Our strategy is inspired by the analogous problem of calculating excitons using the Bethe-Salpeter equation. Excitons can be delocalized over many crystal unit cells, but the underlying Bethe-Salpeter

equation requires only information about DFT Kohn-Sham wavefunctions and Coulomb matrix elements within a single crystal unit cell. In the same spirit, in this work we reformulate the calculation of polarons via DFT as the solution of a coupled non-linear system of equations. The ingredients of such equations are computed in the primitive unit cell using density-functional perturbation theory. By solving these 'polaron equations' we obtain the wavefunction, the lattice deformation, the formation energy, and the spectral decomposition of the self-trapped state into the underlying Bloch states and phonon modes. In order to demonstrate this methodology I will discuss a few case studies including large and small polarons, and show that even in the simplest crystals the structure of polarons is considerably richer than previously thought.

[1] W. H. Sio, C. Verdi, S. Poncé, and F. Giustino, *Phys. Rev. Lett.* **122**, 246403 (2019).

[2] W. H. Sio, C. Verdi, S. Poncé, and F. Giustino, *Phys. Rev. B* **99**, 235139 (2019).

### **Hybrid perovskites: from the random phase approximation to machine learned potentials**

**Georg Kresse**, Menno Bokdam, Ryosuke Jinnouchi

University of Vienna, Vienna - Austria

**Room Orsay, 11:30**

**Wednesday 11**

Reaching predictive accuracy in first principles calculations for complex materials has always been a dream. Here we address this challenge for a very complex highly dynamic material, the hybrid perovskite MAPbI<sub>3</sub> a promising new solar cell material. The first part of the talk addresses the issue, which density functional is the “best” for structure simulations of a particular material? A concise, first principles approach to answer this question is presented [1]. The random phase approximation (RPA) — at least for solids an very accurate many body theory — is used to evaluate and rank various density functionals for MAPbI<sub>3</sub>. The evaluation is done by first creating finite temperature ensembles for small supercells using RPA molecular dynamics, and then evaluating the variance between the RPA and various approximate density functionals for these ensembles. We find that, contrary to recent suggestions, van der Waals functionals do not improve the description of the material, whereas hybrid functionals and the strongly constrained appropriately normed (SCAN) density functional yield very good agreement with the RPA. In the second part of the talk, realistic large scale finite temperature simulations of MAPbI<sub>3</sub> are presented. To achieve the required long simulation times and large length scales, an on-the-fly machine learning scheme that generates force fields automatically during first principles molecular dynamics simulations is used (relying on the previously determined best functional SCAN). This opens up the required time and length scales, while retaining the distinctive chemical precision of first principles methods and minimizing the need for human intervention. Using machine learned potentials, isothermal-isobaric simulations give direct insight into the underlying microscopic mechanisms of the phase transitions. Although used for MAPbI<sub>3</sub> here, the method is widely applicable to multi-element complex systems [2].

[1] M. Bokdam, J. Lahnsteiner, B. Ramberger, T. Schäfer, and G. Kresse, *Phys. Rev. Lett.* **119**, 145501 (2017).

[2] R. Jinnouchi, J. Lahnsteiner, F. Karsai, G. Kresse, and M. Bokdam, *submitted*.

### **DNA a 10<sup>10</sup> multiscale problem**

**Modesto Orozco**

University of Barcelona, Barcelona - Spain

**Room Lausanne, 11:00**

**Wednesday 11**

DNA is not only the “molecule of life”, but also a fascinating molecule with unique properties and one of the most challenging multiscale problems. The study of DNA requires often the analysis of small details, below the Å, but in others, the analysis of the meter-long chromatin fiber is required. Navigating across this multiscale scenario is complex, forcing the use of multi-physics approaches, where different levels of resolution are coupled to different Hamiltonians and different sampling approaches. I will summarize advances done in our group to the development of a continuum of methodologies, from quantum mechanics to mesoscopic models, which allows a holistic and detailed picture of DNA.

### ***Electrode models for computational electrochemistry***

**Mathieu Salanne**<sup>1</sup>, Thomas Dufils<sup>1</sup>, Alessandro Coretti<sup>2</sup>, Michiel Sprik<sup>3</sup>, Laura Scalfi<sup>1</sup>, Benjamin Rotenberg<sup>1</sup>

<sup>1</sup>Sorbonne University, Paris - France

<sup>2</sup>Swiss Federal Institute of Technology (EPFL), Lausanne - Switzerland

<sup>3</sup>Cambridge University, Cambridge United Kingdom

**Room Lyon, 11:30**

**Wednesday 11**

Applied electrochemistry plays a key role in many technologies, such as batteries, fuel cells, supercapacitors or solar cells. It is therefore at the core of many research programs all over the world. Yet, fundamental electrochemical investigations remain scarce. In particular, electrochemistry is among the fields for which the gap between theory and experiment is the largest. From the computational point of view, this is due to the difficulty of combining a realistic representation of the electrode electronic structure and of the electrolyte structure and dynamics. Over the past decade we have developed a classical molecular dynamics code that allows to simulate electrochemical cells. In a first step, the electrodes were modeled as perfectly screening metals with a constant applied potential between them. Recently, we have extended this approach in order to account ; for the degree of metallicity of the electrode (i.e. from semimetals to perfect conductors), using a semi-classical Thomas-Fermi model. In parallel, we have recently shown that it is possible to replace the constant applied potential method by using the finite field method to a system with a slab geometry. The algorithm were also improved in order to reduce the costs of the simulation with respect to conventional classical molecular dynamics. Concerning the applications, these simulations have allowed us to study a variety of electrochemical problems, ranging from the origin of the supercapacitance in nanoporous carbon electrodes to the mechanisms of electron transfer in ionic liquids or molten salts, at the vicinity of planar or porous electrodes.

### ***Self organization and irreversibility in cyclically driven amorphous matter***

**Srikanth Sastry**<sup>1</sup>, Anshul D. S. Parmar<sup>2</sup>, Vinutha H A<sup>3</sup>, Premkumar Leishangthem<sup>4</sup>, Monoj Adhikari<sup>1</sup>, Pallabi Das<sup>1</sup>

<sup>1</sup>Jawaharlal Nehru Centre For Advanced Scientific Research (JNCASR), Bangalore - India

<sup>2</sup>University of Montpellier, Montpellier - France

<sup>3</sup>Chinese Academy of Sciences, Beijing - China

and Cambridge University, Cambridge - United Kingdom

<sup>4</sup>National Institute of Technology, Manipur, India

**Room Blaricum, 11:30**

**Wednesday 11**

Self organization and emergence of irreversibility in externally driven particle assemblies is of interest in many contexts. Specific examples in soft matter systems include the transition from amorphous solid states through the process of yielding and flow, and conversely, shear jamming in granular packings. A variety of such phenomena have been explored by employing cyclic shear deformations or applied stresses, in experiments and computer simulations, and lead to insights in to yielding behaviour of glasses, shear jamming, memory formation, annealing and unjamming in glasses and jammed packings, and the transition from self organized absorbing states to irreversible states. Results from computer simulation investigations of some of these phenomena will be presented, focusing on developing a unified description of transitions from reversible to irreversible behaviour.

### ***Electromechanics of the liquid water-vapour interface***

**Michiel Sprik**<sup>1</sup>, Chao Zhang<sup>2</sup>

<sup>1</sup>University of Cambridge, Cambridge - United Kingdom

<sup>2</sup>Uppsala University, Uppsala - Sweden

**Room Lyon, 10:30**

**Wednesday 11**

The liquid water vapour interface is a classic and enduring molecular simulation topic which has repeatedly featured in Cecam workshops virtually from the early days of Cecam. There is general

agreement that the dipoles of the molecules in the two-molecule deep surface layer are aligned giving rise to a dipole potential. While the symmetry breaking at the interface allows for alignment of dipoles it is somewhat surprising they do because aligning dipoles in a thin sheet costs energy. Even more intriguing is that the value of the computed dipole potential is comparable to the experimental surface potential of 150 mV (going up entering the liquid) now general accepted in the electrochemical literature. The question why remains a challenge for molecular simulation. Most studies, naturally, focus on the structure of the hydrogen bonding at the surface. We have tried to approach the problem from a thermodynamic perspective and have investigated the role of electromechanical coupling. Our SPCE simulations indicate that the surface dipole is related to the response of the surface tension to the application of an electric field normal to the surface. In fact, the MD results suggest that the surface dipole density (within a finite but acceptable error margin) is equal to the zero field derivative of the surface tension. This observation can be rationalised by simple continuum theory. However we are still struggling with the statistical mechanics. After presenting the MD method and results, we will try to summarise our current (still rather limited) understanding about the relevant electromechanics theory.

### ***Ionic liquid confined between metallic interfaces: what is the role of image charges?***

**Samuel Ntim, Marialore Sulpizi**

Johannes Gutenberg University, Mainz - Germany

**Room Lyon, 11:00**

**Wednesday 11**

Peculiar properties of room-temperature ionic liquids in confined geometries have not only become essential for many fields like energy storage, catalysis and tribology, but have also kept puzzling scientists from the fundamental point of view. Recently, an anomalous liquid-solid phase transition has been observed in atomic force microscopy experiments for 1-butyl-3-methylimidazolium tetrafluoroborate ([BMIM][BF<sub>4</sub>]) in different confining materials, the transition being more pronounced for more metallic surfaces. Image charges have been suggested as the key element driving the anomalous freezing. Using classical atomistic molecular dynamics simulations, we systematically investigate here the impact of the image charges on the structure, dynamics and thermodynamics of [BMIM][BF<sub>4</sub>] confined between gold electrodes. Our results not only unveil a minor role played by the metal polarization on the investigated properties, possibly due to the strong electrostatic screening by the ionic liquid, but also provide a novel description of the interfacial layer. We find that, although no diff use layer can be de fined in terms of the electrostatic potential, long range effects are clearly visible in the dynamical properties up to 10 nanometers away from the surface, and are expected to influence viscous forces in the experiments.

### ***The role played by interactions in the assembly of active colloids***

**Chantal Valeriani<sup>1</sup>, Francisco Alarcon<sup>1</sup>, Eloy Navarro<sup>2</sup>, Ignacio Pagonabarraga<sup>3</sup>, Stewart Mallory<sup>4</sup>, Angelo Cacciuto<sup>5</sup>**

<sup>1</sup>Complutense University of Madrid, Madrid - Spain

<sup>2</sup>University of Barcelona, Barcelona - Spain

<sup>3</sup> Swiss Federal Institute of Technology (EPFL), Lausanne - Switzerland

<sup>4</sup>California Institute of Technology (Caltech), Pasadena - USA

<sup>5</sup>Columbia University, New York - USA

**Room Blaricum, 10:30**

**Wednesday 11**

Active matter systems are composed of constituents that consume energy in order to move or exert mechanical forces, constantly driving themselves away from equilibrium [1]. Examples of active particles are living, such as bacteria, or artificial, such as active colloids [2,3]. Experiments on spherical man-made self-propelled colloids have shown that active particles present interesting emergent collective properties [4–6], such as motility-induced phase separation (MIPS), involving spontaneous assembly of particles due to the persistence of their direction of motion [7]. An example of colloids undergoing MIPS under suitable conditions are Active Brownian Particles (ABP), i.e. self-propelled Brownian particles interacting with each other via a purely repulsive potential [8]. In order to design novel functional materials, one might need to gain control on the self-assembly process of active colloids. With this goal in mind, we have explored the competition between activity and a broad range of interactions between active colloids,

ranging from isotropic (strongly repulsive [9,15,17], attractive [10,11], micelleinducing [12]) to anisotropic (Janus-like [13]), unravelling the relevance of hydrodynamics [11,14,16].

- [1] C. Bechinger et al. *Rev. Mod. Phys.* **88**, 045006 (2016).
- [2] W.F. Paxton et al. *Chem. Commun.* **441**, 3 (2005).
- [3] S. Fournier-Bidoz et al. *J. Am. Chem. Soc.* **126**, 13424 (2004).
- [4] S. Thutupalli, R. Seemann, S. Herminghaus *New J. Phys.* **13**, 073021 (2011).
- [5] D. Nishiguchi, Masaki S. *Phys. Rev. E* **92**, 052309 (2015).
- [6] I. Buttinoni, J. Bialké, F. Kümmel, H. Löwen, C. Bechinger, T. Speck. *Phys.Rev. Lett.* **110**, 238301 (2013).
- [7] M.E. Cates, J. Tailleur. *Annu. Rev. of Condens. Matt. Phys.* **6**, pp. 219 (2015).
- [8] S.Mallory, C.Valeriani and A.Cacciuto *Annual Review of Physical Chemistry*, **69** 59 (2018).
- [9] D. Rogel Rodriguez, F. Alarcon, R. Martinez, J. Ramirez, and C. Valeriani, in preparation (2018).
- [10] B. Mognetti, A. Saric, S. Angioletti-Uberti, A. Cacciuto, C. Valeriani and D. Frenkel *Phys.Rev.Lett.* **111**, 245702 (2013)
- [11] F.Alarcon, C.Valeriani and I.Pagonabarraga *Soft Matter* **10**,1039 (2017).
- [12] C.Tung, J.Harder, C.Valeriani and A.Cacciuto, *Soft Matter* **12**, 555 (2016).
- [13] S.Mallory, F.Alarcon, A.Cacciuto and C.Valeriani, *New Journal of Physics* **19**, 125014 (2017).
- [14] F.Alarcon, E.Navarro, C.Valeriani and I.Pagonabarraga, *Phys. Rev. E* **99**, 062602 (2019).
- [15] J.Harder, S.Mallory, C.Tung, C.Valeriani and A.Cacciuto, *J. Chem. Phys.* **141**, 194901 (2014).
- [16] R.Martinez, F.Alarcon, D.R.Rodriguez, J.L.Aragones and C.Valeriani, *EPJE* **41**, 91 (2018).

### ***Biosimulations in life sciences: coarse-grained molecular simulation models are usually superb, but when do we dare to use them?***

**Ilpo Vattulainen**

Tampere University of Technology, Tampere - Finland

Room Lausanne, 10:30

Wednesday 11

Processes in living systems take place over a multitude of different scales in time and place. Therefore, different techniques have been developed to describe quantum-mechanical phenomena where electronic degrees of freedom are important, atomistic phenomena where reactions do not take place but consideration of atomistic details is essential, molecular phenomena where molecular features have to be accounted for but atomistic details are no longer particularly relevant, and continuum-scale phenomena where molecular features no longer matter. Every one of these steps involves coarse graining from a more detailed to a less detailed description, yet in commonly used terminology the concept of “a coarse-grained model” often stands for a molecular description that has been coarse grained from an atomistic one. Here, we follow the same practice and discuss in a life science context one of the quite fundamental questions related to computer simulations in the atomistic and molecular regimes: Under what circumstances can we use coarse-grained molecular models reliably? Unfortunately, the validity of the results of coarse-grained models is still occasionally taken for granted, while real-life observations can be much trickier. We discuss this general problem through a few case studies, some of which originally produced quite surprising results. We first consider protein-protein oligomerization and conclude that paying attention to atomistic details would be reasonable if the aim is to find the correct dimerization interfaces. We move on to discuss tear film lipid layers associated with dry eye syndrome and realize that inclusion of atomistic details in the simulation model helps quite a bit to predict the correct evaporation rates of water across the lipid layer. We further discuss diffusion of membrane proteins and nanoparticle-tagged membrane probes and conclude that in this context atomistic details typically do not matter much. Finally, we briefly consider certain biomolecular features where atomistic accuracy should never be ignored. Altogether, both atomistic and coarse-grained simulation models can be exceptionally good, or exceptionally bad, depending on the context where they are used. Yet, to rely on their results, these case studies demonstrate that they should at least produce the correct phase behavior and describe the necessary chemical details sufficiently well, and obviously it would be positive if their results were consistent with experimental data.



## ***A jamming transition affects landscape and generalisation in deep learning***

**Matthieu Wyart**

Swiss Federal Institute of Technology (EPFL), Lausanne - Switzerland

**Room Blaricum, 11:00**

**Wednesday 11**

Deep learning has been immensely successful at a variety of tasks, ranging from classification to AI. Learning corresponds to fitting training data, which is implemented by descending a very high-dimensional loss function. Understanding under which conditions neural networks do not get stuck in poor minima of the loss, and how the landscape of that loss evolves as depth is increased remains a challenge. Here we predict, and test empirically, an analogy between this landscape and the energy landscape of repulsive ellipses. We argue that in fully-connected networks a phase transition delimits the over- and under-parametrized regimes where fitting can or cannot be achieved. In the vicinity of this transition, properties of the curvature of the minima of the loss are critical. This transition shares direct similarities with the jamming transition by which particles form a disordered solid as the density is increased, which also occurs in certain classes of computational optimization and learning problems such as the perceptron. Our analysis gives a simple explanation as to why poor minima of the loss cannot be encountered in the overparametrized regime. I will discuss how this transition affects performance, and how performance behaves in the over-parametrized regime where nets become very wide.

## **Contributed talks**

### ***Nanobubble nucleation due to surface vibration***

**Saikat Datta, Rohit Pillai, Matthew K. Borg**

School of Engineering, University of Edinburgh, Edinburgh - United Kingdom

**Room Lyon, 12:20**

**Wednesday 11**

Generation and control of surface nanobubbles has got substantial research attention in the recent years due to its increasing importance in various engineering and biomedical applications like surface cleaning [1], formation of surface nanostructures [2], drag reduction [3], and particle trapping inside microchannels [4]. Ultrasonic vibration of a surface adjacent to the liquid is a well-established technique to produce surface bubbles [5]. Existing experimental investigations of vibrating surfaces suggest that the formation of bubble occurs mainly due to cavitation. Moreover, hydrophobic surfaces are identified to be preferable for such bubble formation [6]. Nevertheless, it is also reported that at the nanoscale there is a significant rise in temperature adjacent to a vibrating solid surface leading to nucleation, i.e. boiling [7]. In this work, we employ molecular dynamics simulations to analyse the interplay between the applied vibrational surface forces and the wettability of the solid surface, on bubble nucleation at the nanoscale. It is observed that high frequency and small amplitude of vibration leads to nucleation due to boiling, whereas the large amplitudes of vibrations generate cavitation bubbles. Nucleate boiling tends to occur on hydrophilic surfaces; cavitation, on the other hand, favours hydrophobic substrates. We produce a regime map to identify the mode of nucleation and the preferential wetting nature of the surface, for bubble formation at a specific amplitude and frequency of vibration. The present findings could be a decisive factor for better design of devices involving surface-driven bubble nucleation.

[1] Z. Wu, H. Chen, Y. Dong, H. Mao, J. Sun, S. Chen, V.S. Craig, and J. Hu, *J. Colloid Interface Sci.* **328**, 10 (2018).

[2] F. Karim, E.S. Vasquez, and C. Zhao, *Optics Letters* **43**, 334 (2018).

[3] S.B. Ramiseti, M.K. Borg, D.A. Lockerby, and J.M. Reese, *Phys. Rev. Fluids* **2**, 084003 (2017).

[4] C. Zhao, Y. Xie, Z. Mao, Y. Zhao, J. Rufo, S. Yang, F. Guo, J.D. Mai, and T.J. Huang, *Lab on a Chip*, **14**, 384 (2014).

[5] V. Belova, D.A. Gorin, D.G. Shchukin, and H. Möhwald, *ACS Applied Materials & Interfaces*, **3**, 417 (2011).

[6] Belova-Magri, V., Brochie, A., Cairós, C., Mettin, R. and Möhwald, H., *ACS Applied Materials & Interfaces* **7**, 4100 (2015).

[7] R. Pillai, M.K. Borg, and J.M. Reese, *Phys. Rev. Lett.* **121**, 104502 (2018).

### **Mapping DNA-photosensitizers interaction in and out the nucleosome**

**Elise Dumont**

Ecole Normale Supérieure de Lyon, Lyon - France

**Room Lausanne, 12:00**

**Wednesday 11**

Formation and repair of DNA lesions embraces a rich and combinatorial chemistry, where atomic-scale simulations are most helpful to complement and expand experimental evidences. This biopolymer constitutes a critical testcase at the crossroad of several timely methodological developments of GPU-accelerated MD codes, polarizable force fields and multiscale approaches [1]. I will present our recent efforts to achieve a robust, computationally-driven description of photosensitizers interacting with DNA. Intensive molecular dynamics allow to map the hot spots for DNA lesion induction and delineate rule-of-thumbs for the non-covalent forces that drive sequence recognition [2]. This allows to build DNA-photosensitizer structures, palliating the absence of NMR or X-ray data for labile drugs, and investigate triplet-triplet energy transfer [3], probe new excited-state mechanisms for DNA photostability [4] and rationalize the DNA-dependent photochemistry of DNA drugs [4] owing to QM/MM-MD schemes. The computational efficiency of the MD-then/QM/MM schemes allows to tackle to chemistry within the nucleosome that unveil interactions between DNA defects and histone tails.

[1] (a) C. Zhang et al., *J Chem Theory Comput.* **14**, 2084 (2018).

(b) I. Ivani et al., *Nat. Methods* **13**, 55 (2016).

[2] E. Bignon et al., *Chem. Eur. J.* **23**, 12845 (2017).

[3] E. Dumont et al., *J. Phys. Chem. Lett.* **6**, 576 (2015).

[4] A. Francés-Monerris et al., *Chem. Sci.* **9**, 7902 (2018).

### **Slowing down supercooled liquids by manipulating their local structure**

**Giuseppe Foffi, Susana Marín Aguilar, Henricus H. Wensink, Frank Smallenburg**

Laboratoire de Physique des Solides (LPS), University of Paris-Sud, Orsay - France

**Room Blaricum, 12:20**

**Wednesday 11**

How do we design particles with interactions capable of inducing a more efficient glassy dynamics in supercooled liquids? While it is well understood that glassy dynamics are accompanied by the emergence of long-lived locally favoured structures [1,2], it is not clear how the such structures can be designed to facilitate dynamical arrest. Here, we present a simple directional attractive colloidal model capable of templating the local structure of binary hard-sphere glasses. While discussing its glassy properties, we show that only few specific symmetry results in a dramatic slowing down of the dynamics [3]. Our results open the door to controlling the dynamics of dense glassy systems by selectively promoting specific local structural environments. The extension of these results to traditional isotropic glass former will be also discussed [4].

[1] G. Tarjus, S.A. Kivelson, Z. Nussinov, P. Viot, *J. Phys.: Condens. Matter* **17**, R1143 (2005).

[2] C.P. Royall, S. R. Williams, *Phys. Rep.* **560**, 1 (2015).

[3] S. Marín Aguilar, H. H. Wensink, G. Foffi, F. Smallenburg *arXiv:1812.00764*.

[4] S. Marín Aguilar, H. H. Wensink, G. Foffi, F. Smallenburg, *to be submitted*.

### **Emergence of long-ranged stress and strain correlations in viscoelastic liquids**

**Matthias Fuchs, M. Maier, F. Vogel, A. Zippelius**

Universität Konstanz, Konstanz - Germany

**Room Blaricum, 12:00**

**Wednesday 11**

A theory for the non-local shear stress correlations in viscoelastic liquids is derived from first principles. It explains the emergence of long-ranged elastic correlations in glass starting from the fluid state. Universal far-field correlations arise from the coupling of shear stress to transverse momentum, which is ignored in the classic Maxwell model. To rescue this widely used model, we suggest a generalization in terms of a single relaxation time. This generalized Maxwell model implies a divergent correlation length and correctly accounts for the emergence of elastic stress patterns first discussed by Eshelby [1]. Generalizing to overdamped systems, we discuss the qualitatively different character of Goldstone modes in the stress response of glasses depending on Newtonian versus Langevin dynamics. A diffusive mode dominates

the linear rheological spectra in colloidal glasses [2]. The theory also predicts long-ranged shear strain fluctuations which show an Eshelby-strain pattern, characteristic for elastic response, even in liquids at times longer than the structural relaxation time and after the shear modulus has relaxed. We verify this using microscopy of colloidal and simulations of binary glass-forming systems [3].

[1] M. Maier, A. Zippelius, and M. Fuchs, *Phys. Rev. Lett.* **119**, 265701 (2017).

[2] F. Vogel, A. Zippelius, and M. Fuchs, *EPL* **125**, 68003 (2019).

[3] M. Hassani, E.M. Zirdehi, K. Kok, P. Schall, M. Fuchs and F. Varnik, *EPL* **124**, 18003 (2018).

### **High pressure liquid hydrogen across molecular dissociation**

**Carlo Pierleoni**

DSFC, University of L'Aquila, L'Aquila - Italy

**Room Lyon, 12:00**

**Wednesday 11**

We studied the transformation from molecular-insulating to monatomic-metallic fluid hydrogen upon increasing pressure, by first principle simulations based on both Density Functional Theory (DFT) and Quantum Monte Carlo (QMC) methods [1,2]. Below a critical temperature  $T_c$  (located between 1500K and 3000K), the transition is first order with a discontinuity in the specific volume, a sudden dissociation of the molecules and a discontinuous change in electronic properties like the momentum distribution and the electrical conductivity [3,4,5]. Above the critical point the transformation into the metallic dissociated state is continuous. Optical response of the system is obtained within the Kubo-Greenwood framework of DFT. Agreement with recent experimental results at NIF[6] for reflectivity and absorption is observed. Our analysis suggest a coherent picture for apparently discordant experiments from different methods.

[1] C. Pierleoni, M.A. Morales, G. Rillo, M. Holzmann and D.M. Ceperley, *PNAS* **113**, 4953 (2016).

[2] M.A. Morales, C. Pierleoni, E. Schwegler and D.M. Ceperley, *PNAS* **107**, 12799 (2010).

[3] C. Pierleoni, M. Holzmann and D.M. Ceperley, *Contrib. Plasma Phys.* **58**, 99 (2018).

[4] C. Pierleoni, G. Rillo, D.M. Ceperley and M. Holzmann, *J. Phys.: Conf. Ser.* **1136**, 012005 (2018).

[5] V. Gorelov, C. Pierleoni and D.M. Ceperley, *Contributions to Plasma Physics* (2019); e201800185. <https://doi.org/10.1002/ctpp.201800185>.

[6] G. Rillo, M.A. Morales, D.M. Ceperley and C. Pierleoni, *PNAS* **116**, 9770 (2019).

[6] P. Celliers et al, *Science* **361**, 677 (2018).

### **Towards an atomistic structure of phase-separated protein granules**

**Anton Polyansky**, Alwin Köhler, Bojan Zagrovic

University of Vienna, Vienna - Austria

**Room Lausanne, 12:20**

**Wednesday 11**

Formation of non-membrane-bound, phase-separated cellular organelles such as P-bodies, nucleoli, and stress granules is crucial for the organization of matter in the living cells and their normal function and is currently receiving a growing interest of a multidisciplinary research community. However, the fundamental physicochemical principles behind such liquid-liquid phase separation, which in the cellular environment has been observed for proteins, RNA and their mixtures, are far from being understood. In fact, elucidation of such principles will likely require development of an advanced theoretical foundation combining molecular biology, biophysics, polymer physics and physical chemistry. Here, we present to our knowledge the first attempt to model self-association of phase-separating, intrinsically disordered proteins at all-atom resolution in an explicit solvent. For this purpose, we choose a set of disordered protein fragments, which have been experimentally characterized as having different propensities to form phase-separated granules. To model such systems, we developed a protocol that utilizes a water model especially developed for molecular dynamics simulation of disordered proteins (TIP4PD). We have simulated model peptides in the microsecond range using different numbers of protein copies corresponding to either an infinite dilution or a highly concentrated protein solution (approx. 10 mM). In agreement with experimental data, we reproduce different behavior of the polypeptides when it comes to dynamics and degree of self-association (multivalency parameter), as well as identify the key types of protein-protein interactions contributing to phase separation. We show that during simulations the interacting peptides form fisherman's net-like structures, where the formation of clusters of interacting molecules reduces protein dynamics and results in stable organization. On the other hand, the growth of clusters depends on the multivalency of protein-protein interactions and may require crossing of a threshold value with  $>2$  simultaneously interacting partners. Importantly, we observe that the modeled

disordered protein fragments display virtually identical modes of intra and intermolecular interactions, suggesting that the network topology and dynamics may be directly encoded in the protein sequence and represent a projection of the intra-molecular interaction patterns in a multiprotein context.

### ***Selective conduction of organic molecules on free-standing graphene and carbon nanotubes***

**Zhao Wang**

Vienna University of Technology, Vienna - Austria and Guangxi University, Nanning - China

**Room Orsay, 12:20**

**Wednesday 11**

Directional transport of molecules via nanostructures holds great promise for a wide range of applications in chemistry and biology. We study the transport of simple organic molecules on free-standing graphene and carbon nanotube substrates driven by thermal gradients via molecular dynamics. Fast conduction of the molecules is observed with selectivity for aromatic compounds. This selectivity stems from the fact that the planar structure of the aromatic molecule helps keep a shorter distance to the substrate, which is the key to the driving force at the gas-solid interface. The drift velocity monotonically increases with decreasing molecule density, with no ballistic transport observable even for a single molecule. A non-linear regime is discovered for the conduction of benzene molecules under large thermal gradients. At low temperature, molecules formed aggregation and move collectively along specific path in the graphene substrate. These results have strong implications for our understanding of the conduction of organic molecules on low-dimensional nanostructures, and pave a way to a practical mean for selectively recognizing planar aromatic molecules, benzene in particular.

### ***Calculating ground state energy surfaces for $H_2O$ and $HcN$ with a quantum computer***

**Göran Wendin**, Mårten Skogh, Laura Garcia Alvarez, Martin Rahm

Chalmers University of Technology, Gothenburg - Sweden

**Room Orsay, 12:00**

**Wednesday 11**

Calculation of the structure and dynamics of interacting many-electron systems is a hard problem that cannot be done efficiently by classical computers. Quantum computers can in principle provide exponential speedup - quantum advantage. Therefore, the calculation of molecular energy surfaces is an important application for present and future quantum computers. To calculate molecular binding energies, the second-quantised Hamiltonian is first transformed to a Pauli matrix spin representation, convenient for implementation on a qubit processor. The energy can then be estimated either by (i) evolving a zeroth order state function in time and applying a phase-estimation algorithm; or (ii) by using the variational quantum eigensolver (VQE) to optimise a trial state function to calculate the expectation value of the Hamiltonian. The VQE is an example of a hybrid scheme where the trial state function is set up and the energy is calculated in the quantum processor, while the optimisation of the parameters in the trial function is done by the classical processor. In this work we use the VQE to calculate parts of the ground state energy surfaces of  $H_2O$  (12 qubits; basis) and  $HcN$  (16 qubits; basis) with the QISKit-aqua-chemistry software package using minimal basis sets. The quantum parts of the algorithm is run on a HPC backend simulating a quantum computer - there are currently no 12-16 quantum processors with coherence time long enough to calculate the Hamiltonian averages with sufficient accuracy. Nevertheless, applications to quantum chemistry will be important for benchmarking noisy intermediate-scale quantum processors (NISQ).

## Plenary speaker

### ***Non-unitary and stochastic quantum chemistry***

**Ali Alavi**

Max Planck Institute for Solid State Research, Stuttgart - Germany

Room Lausanne, 09:00

Thursday 12

Efficient methods to solve the electronic Schrodinger equation require a means to incorporate electron-electron distances and yet more sophisticated variables into the description of the wavefunction. Such explicit correlation methods are usually extremely complicated, and in addition, in their present formulations in quantum chemistry are adapted only for weakly correlated systems. Here we investigate non-unitary similarity transformations of the Hamiltonian, which transform such explicitly correlated variables in the Hamiltonian, resulting in non-hermitian problems. We show that the imaginary time dynamics of projective Quantum Monte Carlo methods are ideally suited to solve such non-hermitian problems, and provide a natural framework for the study of strongly correlated electronic systems as well as largely solving the basis set problems of ab initio quantum chemistry. There are complications, however: the resulting Hamiltonians, in addition to be non-hermitian, also contain three-electron interactions. We show that stochastic algorithms, in which such terms are sampled (rather than deterministically evaluated) provide a means forward, although challenges remain to scaling up to large systems. Perspectives on future directions will be discussed.

### ***Molecular dynamics simulations in the age of machine learning***

**Eric Vanden-Eijnden**

Courant Institute, New York University, New York - USA

Room Lausanne, 12:25

Thursday 12

The rapid developments in machine learning (ML), leading to its success in the context of complex classification tasks, offer intriguing promises for molecular dynamics simulations. Indeed, ML has already been successfully used for force field parametrization, protein structure prediction, drug design, etc. These tasks rely on processing data obtained by independent means. Here I will discuss how ML could also help in producing long time series data, specifically focusing on free energy calculations and reactive event analysis, two problems that require designing accelerated sampling strategies and can possibly lead to learning low dimensional models.

## Invited speakers

### ***Quantum and classical ripples in graphene***

**Roman Martonak**<sup>1</sup>, Juraj Hašík<sup>1,2</sup>, Erio Tosatti<sup>2,3,4</sup>

<sup>1</sup>Comenius University in Bratislava, Bratislava - Slovakia

<sup>2</sup>Scuola Internazionale Superiore di Studi Avanzati (SISSA), Trieste - Italy

<sup>3</sup>CNR-IOM Democritos, Trieste - Italy

<sup>4</sup>International Centre for Theoretical Physics (ICTP), Trieste - Italy

Room Orsay, 10:30

Thursday 12

Thermal ripples of graphene are well understood at room temperature, but their quantum counterpart at low temperatures are in need of a realistic quantitative description. Here we present atomistic Path Integral Monte Carlo (PIMC) simulations of freestanding graphene, which show upon cooling a striking

classical-quantum evolution of height and angular fluctuations [1]. The crossover takes place at ever-decreasing temperatures for ever-increasing wavelengths so that a completely quantum regime is never attained. Zero-temperature quantum graphene is flatter and smoother than classical graphene at large scales, yet rougher at short scales. The angular fluctuation distribution of the normals can be quantitatively described by coexistence of two Gaussians, one classical, strongly T-dependent, and one quantum, about 2° wide, of zero-point character. The quantum evolution of ripple-induced height and angular spread should be observable in electron diffraction in graphene and other 2D materials like MoS<sub>2</sub>, bilayer graphene, BN, etc.

[1] J. Hašík, E. Tosatti and R. Martoňák, *Phys. Rev. B* **97**, 140301(R) (2018).

**Unified theory of thermal transport in crystalline solids and glasses.**  
**Francesco Mauri**

University of Rome la Sapienza, Rome - Italy

**Room Blaricum, 11:00**

**Thursday 12**

Thermal conductivity in crystalline solids with well-separated phonon branches (namely with few atoms per unit cell) is well describes by a particle-like Boltzmann Equation for phonons, where the main source of scattering is anharmonicity. In such systems the conductivity decreases with temperature (T), with a 1/T law for T larger than the Debye Temperature (T<sub>D</sub>). In amorphous solids the main source of scattering is the static and T-independent disorder, well described by the harmonic theory introduced by Allen and Feldman. In these systems the conductivity increases with temperature and, within the Allen and Feldman theory, reaches a constant value for T>T<sub>D</sub>. Finally periodic solids with large super-cells and very low thermal conductivity can exhibit a glass-like behavior, that can not be described by existing theories. Anharmonicity or disorder are the limiting factors for thermal conductivity in crystals or glasses; hitherto, no transport equation has been able to account for both. Here, we derive such equation, resulting in a thermal conductivity that reduces to the Peierls and Allen-Feldman limits, respectively, in anharmonic crystals or harmonic glasses, while also covering the intermediate regimes where both effects are relevant [1]. This approach also solves the long-standing problem of accurately predicting the thermal properties of crystals with ultralow or glass-like thermal conductivity, as we show with an application to a thermoelectric material representative of this class.

[1] M. Simoncelli, N. Marzari, F. Mauri, *Nature Physics* **15**, 809 (2019).

**Multiscale simulations of nanofluidic porous systems for eenergy applications**  
**Simone Meloni**

University of Ferrara, Ferrara - Italy

**Room Lausanne, 10:30**

**Thursday 12**

*Nanoturisation*, the shrinking of the characteristic size of devices and materials to the nanoscale, is changing the perspective of fundamental and applied sciences. In engineering, and fluidics in particular, nanoturisation completely changed the perspective as effects that have often been discarded, namely thermal fluctuations, became key phenomena to take into account. Thus, methods and theories developed in molecular physics and chemistry, such as rare events methods, are *percolating* to nanoengineering and nanofluidics. In this contribution I will talk about a phenomenon relevant to nanofluidics: intrusion and extrusion of liquids from nanoporous materials and nanotextured surfaces. Intrusion and extrusion of liquids from lyophobic (non-wettable) porous materials and textured surfaces are important processes to develop devices for storing mechanical energy, dampening vibrations or absorbing crashes: during intrusion the energy injected into the system is transformed into interface energy, and along extrusion this energy is either returned or dissipated as heat. For which application a material is suitable for depends on the degree of hysteresis, the difference between the intrusion and extrusion pressure: materials with no or minimal hysteresis are suitable for energy storage (no energy is wasted during the active, discharge cycle), systems with large or infinite (no-extruding materials) hysteresis are optimal for dampening or crash absorption. Intrusion/extrusion hysteresis is associated to the presence of wetting/dewetting barriers that, like in any thermally activated event, determines the kinetic of the process, allowing or preventing the spontaneous process over the relevant - experimental or operational - timescale. Thus, understanding the relation between the chemistry and morphology of

porous systems and the wetting/dewetting barrier is key to design novel materials better suited for either class of nanoengineering applications.

In this talk I will revise recent progresses in the field; I will show how simulations and experiments are contributing synergically to develop this research field at the frontier between molecular sciences and engineering, and I will illustrate novel methodological challenges posed by nanofluidics.

### ***Coarse-graining out of equilibrium***

**Tanja Schilling**

University of Freiburg, Freiburg - Germany

**Room Lyon, 11:00**

**Thursday 12**

Complex microscopic many-body processes are often interpreted in terms of "reaction coordinates", i.e. in terms of the evolution of a small set of coarse-grained, ensemble averaged variables. Under stationary conditions, the evolution of such coordinates is described by the generalized Langevin equation. In contrast, if the dynamics is not stationary, it is not a priori clear which form the equation of motion for an averaged observable has. We employ the formalism of time-dependent projection operator techniques to derive the equation of motion for a non-equilibrium trajectory-averaged observable as well as for its autocorrelation function. We consider, in particular, Hamiltonians and observables which depend on time explicitly as e.g. in systems under external driving. The equation of motion which we obtain is similar in structure to the generalized Langevin equation, but it exhibits a time-dependent memory kernel as well as a fluctuating force that implicitly depends on the initial conditions of the process. We also derive a relation between this memory kernel and the autocorrelation function of the fluctuating force that has a structure similar to a fluctuation-dissipation relation. In addition, we show how the choice of the projection operator allows to relate the Taylor expansion of the memory kernel to data that is accessible in MD simulations and experiments, thus allowing to construct the equation of motion. As a numerical example, the procedure is then applied to crystal nucleation from a supercooled Lennard-Jones melt.

### ***Atomistic and mesoscopic simulations of polymer melts***

**Doros Theodorou**

National Technical University, Athens - Greece

**Room Blaricum, 10:30**

**Thursday 12**

The characteristic lengths and times governing structure and molecular motion in polymer melts span many orders of magnitude; as a consequence, predicting their properties defies conventional atomistic simulation techniques. Fortunately, one can develop systematic statistical mechanics-based approaches that meet the challenge of long time and length scales. We will discuss a strategy based on atomistic and mesoscopic hybrid particle/field simulations for the calculation of structure, thermodynamic, and dynamical properties of long-chain, entangled polymer melts in the bulk and at interfaces. The atomistic and mesoscopic levels are closely coupled, the former providing effective force field expressions and parameters for the latter. We start by thoroughly equilibrating the atomistic model using connectivity-altering Monte Carlo simulations, probing its entanglement structure by topological analysis, and interrogating its dynamics for time scales up to microseconds by equilibrium molecular dynamics. In the mesoscopic model, chain segments of ca. 50 skeletal atoms are mapped into single interaction sites or "beads" [1]. Effective bonded interactions for pairs and triplets of successive beads are obtained by Boltzmann-inverting the corresponding distribution functions accumulated in the course of the atomistic simulations. Entanglements at the mesoscopic level are represented through slip-springs connecting different chains and capable of hopping between successive beads along a chain contour, this hopping motion being viewed as an infrequent event [1,2]. The mesoscopic model is grand canonical with respect to the entanglements, with slip-springs being created and destroyed at chain ends and their mean density in the polymer being controlled through a preset "activity" [1]. Nonbonded interactions, on the other hand, are described through an excess Helmholtz energy that is computed from the local density everywhere in the system via an equation of state. The time evolution of the mesoscopic model is tracked by Brownian Dynamics/kinetic Monte Carlo (BD/kMC) simulations [1,2]. Brownian dynamics in the high friction limit displaces the beads subject to the systematic forces generated from the mesoscopic Helmholtz energy function, with a friction constant computed from atomistic molecular dynamics. In parallel, Kinetic Monte Carlo tracks elementary events of slip-spring hopping along chain contours, slip-spring destruction and

creation at chain ends, and bead sorption/desorption at solid surfaces, if present. Rate constants for the processes of sorption and desorption are extracted by hazard analysis of atomistic molecular dynamics trajectories. We apply the mesoscopic model to polyethylene and cis-1,4 polyisoprene melts at equilibrium and subject to shear flow (simulated through Lees-Edwards boundary conditions) at various rates. The zero-shear viscosity, the chain self-diffusion coefficient, and the storage and loss moduli of the melt are predicted in excellent agreement with atomistic simulations and experiment [1,2]. Furthermore, under nonequilibrium (shear flow) conditions, nonlinear viscoelastic properties (shear thinning, first and second normal stress differences) are correctly predicted and explained in terms of changes in the orientation, conformation, and state of entanglement of the chains [1]. The mesoscopic model is 2 to 3 orders of magnitude faster than atomistic molecular dynamics and thus greatly extends the range of time scales and chain lengths that can be studied.

Application of the mesoscopic BD/kMC approach to polymer melt surfaces is achieved by introducing a square density gradient contribution to the Helmholtz energy of nonbonded interactions [3]. The mesoscopic simulations can capture the chain length dependence of surface tension, changes in overall chain shape, size and orientation induced by the surface, the loss of entanglements, and the accelerated dynamics in the free surface region in excellent agreement with atomistic simulations and experiments [3,4]. Furthermore, application of the mesoscopic BD/kMC approach to graphite-molten polyethylene interfaces yields reasonable values for the adhesion tension and elucidates the residence time of chains adsorbed from the melt on the solid substrate as a function of chain length.

[1] A.P. Sgouros, G. Megariotis, D.N. Theodorou, *Macromolecules* **50**, 4524 (2017).

[2] G.G. Vogiatzis, G. Megariotis, and D.N. Theodorou, *Macromolecules* **50**, 3004 (2017).

[3] A.P. Sgouros, A.T. Lakkas, G. Megariotis, D.N. Theodorou, *Macromolecules* **51**, 9798 (2018).

[4] A.P. Sgouros, G.G. Vogiatzis, G. Kritikos, A. Boziki, A. Nikolakopoulou, D. Liveris, D.N. Theodorou, *Macromolecules* **50**, 8827 (2017).

### ***Impact of wall constraint on the dynamics of self-propelled particles***

**Ryoichi Yamamoto<sup>1</sup>, John J. Molina<sup>1</sup>, Federico Fadda<sup>1</sup>, Shun Imamura<sup>2</sup>**

<sup>1</sup>University of Kyoto, Kyoto - Japan

<sup>2</sup>Tohoku University, Sendai - Japan

**Room Lausanne, 11:00**

**Thursday 12**

The presence of wall constraint strongly affects the motions of dispersed particles in a fluid. A striking example can be seen in the dynamics of self-propelled particles near fluid/solid boundaries where the single (collective) motion of such particle(s) depend sensitively on the detailed flow profile around them. In the present study, we investigate the dynamics of two popular types of self-propelled particles, i.e., spherical micro-swimmers (squirmers) and rolling spheres (Quincke rollers) on a flat plate and by means of direct numerical simulation of fluid/particle composite systems with fully resolving the hydrodynamics.

## **Contributed talks**

### ***The maximum probability domains (MPD) as a powerful tools for analysing electronic structures.***

**Mauro Causà**

Università di Napoli Federico II, Napoli - Italy

**Room Orsay, 11:30**

**Thursday 12**

Any tool for interpreting the electronic structures, in stationary or dynamic chemical phenomena, are based on a variable amount of arbitrariness: let think to Mulliken charge analysis, depending on computational features like basis set, but extremely valuable in a first measurement of the electronic charge polarization. The Electronic Localization Function (ELF) is extremely valuable in recognizing the physical basis of chemical phenomenological concept, like the electron pair. The Maximum Probability Domain (MPD) have been recently introduced by Andreas Savin [1]. The MPDs limit the arbitrariness in their definition to the well defined quantum probability, and allow to define and compare "a priori" chemical



phenomenological situation, recognizing or not their physical background. Condensed matter presents very peculiar and extreme situation, like complete electronic localization like in covalent and very ionic materials, and complete electronic delocalization, like in metals. The present survey of MPD analysis opens perspectives to the analysis of electronic structures during reaction events. Examples will be given about applications of MPD analysis in coordination-chemistry and variable-valence compounds [2].

[1] E. Cancès, R. Keriven, F. Lodier, A. Savin, *Theor. Chem. Acc.* **111**, 373 (2004).

[2] M. Causà, A. Savin, *Z. Anorg. Allg. Chem.* **637**, 882 (2011).

### **Machine learning for atomic and molecular simulations**

**Michele Ceriotti**

Swiss Federal Institute of Technology (EPFL), Lausanne - Switzerland

**Room Lyon, 10:30**

**Thursday 12**

Machine learning is finding applications to more and more tasks, in science as much as in everyday life. In this talk I will focus on how atomic and molecular simulations are being transformed by the use of statistical regression models, that make it possible to approximate accurately and efficiently atomistic properties computed from a few reference electronic-structure calculations. I will argue about the advantages that are brought about by a physically-motivated framework, and about the insights that can be obtained by a critical application of ML methods. Examples will be given spanning molecular and condensed matter systems, and properties as diverse as magnetic nuclear chemical shieldings and the electron charge density, underscoring the general applicability of the process.

### **Open boundaries in hamiltonian adaptive resolution simulations: from grand canonical to nonequilibrium molecular dynamics**

**Robinson Cortes Huerto<sup>1</sup>, Maziar Heidari<sup>1</sup>, Kurt Kremer<sup>1</sup>, Raffaello Potestio<sup>2</sup>**

<sup>1</sup>Max Planck Institute for Polymer Research, Mainz - Germany

<sup>2</sup>University of Trento, Trento - Italy

**Room Lyon, 11:50**

**Thursday 12**

The adaptive resolution technique (AdResS/H-AdResS) [1], where atomistic and coarse grained representations of the same system coexist in thermodynamic equilibrium, enjoys a long history of successful applications as a multiscale tool to investigate various challenging problems in soft matter [2]. In a new light, we have recently introduced the Spatially-Resolved Thermodynamic Integration (SPARTIAN) method [3], where the coarse grained model has been replaced by a reservoir of non-interacting particles, and the free energy compensations needed to guarantee a constant density for the whole system have been computed and identified with the excess chemical potential. Indeed, SPARTIAN is a new tool aiming at computing absolute free energies of complex molecular systems.

In this talk, I will guide you through the SPARTIAN technique and illustrate its advantages and drawbacks with examples that include the calculation of chemical potentials, free energies of solids and solvation free energies. Furthermore, since particles do not interact in the reservoir, it is straightforward to insert/delete particles to control the chemical potential of the system. Therefore, I will conclude with an application of this grand canonical molecular dynamics technique to the study of nonequilibrium processes at the nanoscale.

[1] Praprotnik et. al., *J. Chem. Phys.* **123**, 224106 (2005); Potestio et. al., *Phys. Rev. Lett.* **110**, 108301 (2013).

[2] Praprotnik et. al., *Handbook of Materials Modeling: Methods, theory and modeling*, 1 (2018).

[3] Heidari et. al., *J. Chem. Theory Comput.* **14**, 3409 (2018).

***Stabilization of agl's polar surfaces by the aqueous environment, and its implications for ice formation***

**Stephen J. Cox**, Thomas Sayer

University of Cambridge, Cambridge - United Kingdom

**Room Blaricum, 11:30**

**Thursday 12**

Silver iodide is one of the most potent inorganic ice nucleating particles known, a feature generally attributed to the excellent lattice match between its basal Ag-(0001) and I-(000-1) surfaces, and ice. This crystal termination, however, is a type-III polar surface, and its surface energy therefore diverges with crystal size unless a polarity compensation mechanism prevails. In this simulation study, we investigate to what extent the surrounding aqueous environment is able to provide such polarity compensation. On its own, we find that pure H<sub>2</sub>O is unable to stabilize the AgI crystal in a physically reasonable manner, and that mobile charge carriers such as dissolved ions, are essential. In other words, proximate dissolved ions must be considered an integral part of the heterogeneous ice formation mechanism. The simulations we perform utilize recent advances in simulation methodology in which appropriate electric and electric displacement fields are imposed. A useful by-product of this study is the direct comparison to the commonly used Yeh-Berkowitz method that this enables. Here we find that naive application of the latter leads to physically unreasonable results, and greatly influences the structure of H<sub>2</sub>O in the contact layer. We therefore expect these results to be of general importance to those studying polar/charged surfaces in aqueous environments.

***Maximum likelihood analysis of reaction coordinates during crystal nucleation in Ni***

**Grisell Diaz Leines**, Jutta Rogal

Interdisciplinary Centre for Advanced Materials Simulation, Ruhr-University Bochum, Bochum - Germany

**Room Lyon, 11:30**

**Thursday 12**

Fundamental knowledge of the mechanism of crystal nucleation and polymorph selection is essential for the synthesis and control of materials with targeted properties. However, understanding the atomistic mechanism of crystallization poses a major challenge as many materials exhibit complex transitions of multiple steps, forming polymorphic structures. In this work we employ transition path sampling simulations together with a maximum likelihood estimation (MLE) and perform a quantitative and multi-dimensional analysis of the reaction coordinate (RC) and the free energy landscape of the nucleation process in Ni. The MLE approach allows us to directly compare the quality of different order parameters as RCs based on their ability to model the committor function with respect to the given data of the path ensemble. The analysis of the reweighted path ensemble and the free energy surface on a set of candidate structural order parameters shows that the nucleation pathway in Ni is governed by the initial formation of mesocrystal regions and a subsequent emergence of FCC-HCP crystallites embedded within the core of these prestructured clusters. Our findings indicate that the preordered liquid region is an order parameter that enhances the RC and therefore has an essential role in the structural description of the nucleus and its interfacial free energy. Moreover, we show that these regions of higher order than the liquid predetermine the coordination of the FCC-HCP polymorphs selected, acting as precursors of the crystallization. These results shed light on the prominent role of the prestructured liquid regions during crystallization and polymorph selection in Ni.

***Path integral molecular dynamics for cold bosons***

**Barak Hirshberg**, Valerio Rizzi, Michele Parrinello

ETH Zürich, Zürich - Switzerland

**Room Lausanne, 11:30**

**Thursday 12**

Cold trapped atoms are fascinating systems which exhibit fundamental physical phenomena but can also be potentially used in various technological applications. We present a new method for simulating cold Bosons using path integral molecular dynamics. The main difficulty in performing such simulations is enumerating all ring-polymer configurations, which arise due to permutations of identical particles. We show that the potential and forces at each time step can be evaluated using a recurrence relation which

avoids enumerating all permutations, while providing the correct thermal expectation values. The resulting algorithm scales cubically with system size. The method is tested and applied to Bosons in a 2D trap and agrees with analytical results and numerical diagonalization of the many-body Hamiltonian. An analysis of the role of exchange effects at different temperatures, through the relative probability of different ring-polymer configurations, is also presented.

### **Accessing the accuracy of density functional theory through structure and dynamics of the water–air interface**

Tatsuhiko Ohto<sup>1</sup>, Mayank Dodia<sup>2</sup>, Yuki Nagata<sup>2</sup>

<sup>1</sup>Osaka University, Osaka - Japan

<sup>2</sup>Max Planck Institute for Polymer Research, Mainz - Germany

**Room Orsay, 11:50**

**Thursday 12**

Density functional theory (DFT)-based molecular dynamics simulations have been increasingly used for simulating water-gas and water-solid interfaces, while choice of the appropriate exchange-correlation (XC)-functional is still arbitrary due to dissension for the recommended functional. Here, we assess the performance of generalized gradient approximation (GGA), meta-GGA, and hybrid-GGA XC functionals, based on the metrics relevant to sum-frequency generation spectroscopy of water at the water-air interface. The structure and dynamics of water at the water-air interface is governed by heterogeneous intermolecular interactions, providing a critical benchmark for XC-functionals. Understanding on the performance for XC-functionals can resolve the controversial interpretation of the structure at the water interfaces and direct the design of novel XC-functionals to describe the heterogeneous interactions accurately in condensed phases.

[1] T. Ohto et al., *Journal of Chemical Theory and Computation* **15**, 595 (2019).

[2] M. Dodiya, T. Ohto et al, *Journal of Chemical Theory and Computation*, in press.

[3] T. Ohto et al., in preparation.

### **Monitoring water splitting at graphene edges: insights into the effects of humidity on the lubricity of graphitic materials**

Clelia Righi, Paolo Restuccia, **Mauro Ferrario**

Department of Physics, Informatics and Mathematics, University of Modena and Reggio Emilia, Modena - Italy

**Room Orsay, 11:00**

**Thursday 12**

One of the main difficulties in understanding friction is related to the complexity and variety of phenomena that take place at the buried interface, which is not easily accessible by experiments. Simulations can, thus, play a crucial role in this context. In particular, those based on quantum mechanics, which is essential to accurately describe the behavior of the material in conditions of enhanced reactivity as those imposed by the mechanical stresses applied. We apply a quantum-mechanics/molecular-mechanics (QM/MM) approach to monitor in real time a sliding interface containing graphene ribbons and water molecules. The simulations reveal that water intercalation is not expected to survive under the effects of an applied load and it can be definitely ruled out as the origin of graphite lubricity. On the contrary, the simulations point at the key role of tribochemical reactions occurring at graphene edges. The importance of humidity in quenching the reactivity of the graphene edges is highlighted along with the cooperative mechanisms, such as Grotthus-like proton diffusion, that are enhanced by confinement. Furthermore, we quantify the dramatic change in the hydrophilic character of graphene induced by the presence of passivated edges and the attractive interaction arising among them. The importance of the different observed mechanisms in providing graphite lubricity in humid environments is discussed.

### ***Oxidation products of polyethylene and their optical signatures***

**Guido ROMA<sup>1</sup>**, Fabien Bruneval<sup>1</sup>, Layla Martin-Samos<sup>2</sup>, Yunho Ahn<sup>1</sup>

<sup>1</sup>CEA, DEN, Service de Recherches de Métallurgie Physique, Université Paris-Saclay, Gif sur Yvette - France

<sup>2</sup>CNR-IOM DEMOCRITOS, Istituto Officina dei Materiali, c/o SISSA Scuola Internazionale Superiore di Studi Avanzati, Trieste - Italy

**Room Blaricum, 11:50**

**Thursday 12**

Polyethylene (PE), one of the simplest and most used aliphatic polymers, is generally provided with a number of additives, in particular antioxidants, because of its tendency to get oxidized. Oxidation kinetics of PE considered to proceed through a variety of reactions [1] involving several radical species and chain defects: peroxy radicals, hydroperoxides, carbonyl groups ---in the form of ketones, where C=O double bond substitutes a CH<sub>2</sub> group, or aldehydes (chain-end defects)---, carboxylic acids, C=C double bonds, and some combinations thereof. Experimentally, the titration of carbonyl defects using infrared spectroscopy is the main tool to follow the oxidation, or the radio-oxidation, of the material. However, coupling it to optical probes would help to understand in more detail the kinetic mechanisms of radio-oxidation and to link them to the observed chemiluminescence. Many experimental investigations of the optical properties in the visible/UV range mainly attribute the photoluminescence of PE to one specific kind of unsaturated carbonyls. However, the reason why other species, e.g. saturated carbonyls like ketones, are generally overlooked is not clear. We use Density Functional Theory to investigate the stability and some relevant kinetic barriers associated to the formation of carbonyl defects, and we investigate the optical properties of PE containing defects using perturbative GW and the Bethe-Salpeter equation in order to take into account excitonic effects[2]. We compare the calculated excitonic states with experimental absorption/emission spectra and discuss them in the light of the stability of some relevant chain defects.

[1] M. Da Cruz et al., *J. Appl. Polym. Sci.* **133**, 43287 (2016).

[2] G. Roma, F. Bruneval, L. Martin-Samos, *J. Phys. Chem. B* **122**, 2023 (2018).

### ***Towards free-energy profiles for nano-catalyzed chemical reactions in complex environments***

**Dennis R. Salahub**

University of Calgary, Calgary - Canada

**Room Lausanne, 11:50**

**Thursday 12**

I will review our attempts to build somewhat realistic models of nanocatalysis at finite temperature. Current thoughts are to bring in machine-learning techniques to, ideally, define the relevant reaction coordinates/collective variables. Significant progress has been made on such questions in the bio-modeling literature and I would like to understand the new ML methodologies better and to, hopefully, adapt them to the field of nanocatalysis. I am a neophyte, eager for any guidance that CECAM participants might offer, once I have exposed my state of knowledge/ignorance. I will report on progress.

## **Posters**

### ***Nucleation of lead halide perovskites***

**Paramvir Ahlawat<sup>1</sup>, Michele Parrinello<sup>2</sup>, Ursula Rothlisberger<sup>1</sup>**

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Perovskite solar cells have reached remarkable efficiencies of 24.2% [1]. Control over morphology plays a very important role in obtaining high efficiencies of lead halide perovskites solar cells. Nucleation and crystal growth processes dictate the evolution of morphology. To probe the atomic level mechanism of nucleation, experimental methods are limited by spatial and temporal resolution. Therefore, we perform molecular dynamics (MD) simulations of homogeneous nucleation of lead halide perovskites. However, nucleation from solution is a typical example of a rare event process and the time scales are not affordable for standard MD simulations. Here, we use the enhanced sampling technique of well-tempered Metadynamics to overcome these challenges. Metadynamics employs a bias potential which is constructed from a few collective coordinates of the system. In this study, we introduce a new generalized reaction coordinate to enhance the fluctuations. From our simulations [2], we layout the individual stages of the nucleation of lead halide perovskites as depicted in Figure 1. We find that the monovalent cations plays a very important role to initiate the nucleation process. We also identify the relevant intermediate metastable structures formed during nucleation. Our simulations reveal the in-depth details of the nucleation process that helped us to design new experiments to fabricate better perovskite optoelectronics. Spin-coating is the most used technique to make perovskite solar cells. Homogeneous nucleation from solution is a widely used method to make perovskite single crystals. Based on our simulations, we demonstrate the impact of our simulations from new experiments both on spin-coating and homogeneous nucleation. Moreover, perovskites and nucleation is an ubiquitous process in natural as well as in many industrial processes. Our simulations and methods also contribute to the fundamental understanding of nucleation.

[1] <https://www.nrel.gov/pv/cell-efficiency.html>.

[2] P. Ahlawat, P. Piaggi, K. Graetzel, M. Parrinello, U. Rothlisberger, *arXiv preprint arXiv:1810.00759* (2018).

### ***Theoretical and experimental study of new hybrid compound 2-carboxyanilinium hypophosphite***

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Studies of organic-inorganic hybrid materials, including aminoacids and various inorganic acids [1,2], have received a great deal of attention in recent years, because of their electrical, magnetic and optical properties[3]. Hydrogen bonds in hybrid compounds are of interest because of their widespread biological occurrence. The potential importance of hydrogen bonding in the structure and function of biomolecules is well established [4]. A novel organic-inorganic hybrid material 2-carboxyanilinium hypophosphite was synthesized by slow evaporation. The crystal structure was determined by X-ray at room temperature in the centrosymmetric triclinic space group P-1. The X-ray crystal structure of the title compound ( $C_7H_8NO_2^+ \cdot H_2PO_2^-$ ) show extensive stacking interactions, the anions and cations were associated via hydrogen bonding giving a one-dimensional network structure. In the former crystal, the cations, anions molecules are linked into layers by two types of hydrogen bonds: N-H...O and O-H...O. The optimized geometric parameters, normal mode frequencies and corresponding vibrational assignments of the present compound were theoretically examined by the DFT/B3LYP method with the 6-31G (d,p) and 6-311G (d,p) basis sets. The FT-Raman and FT-IR spectra of the compound were determined and compared to the theoretical spectra. The results revealed that the deviation of the theoretical wavenumbers from the experimental values was very small.

[1] N. Benali-Cherif, L. Bendheif, K. Bouchouit, A. Cherouana, H. Merazig *Ann. Chim. Sci. Mat.* **29**, 11 (2004).

[2] L. Bendheif, N. Benali-Cherif, L. Benguedouar, K. Bouchouit, *Acta Cryst. E* **59**, o141(2003).

[3] C.R. Kagan, D.B. Mitzi, C.D. Dimitrakopoulos, *Science* **286**, 945(1999).

[4] G.A. Jeffrey, W. Saenger W., *An Introduction to Hydrogen Bonding*, Springer Verlag, Berlin, 245,(1991).

## **Multiscale approaches for the accurate prediction of protein-ligand binding**

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Protein-ligand interactions are crucial for many important biological processes and thus the selective inhibition of disease relevant proteins is one of the major strategies for drug design in pharmaceutical industry. In this context computer-based methods are routinely used to screen large compound libraries for the identification of potential drug candidates. Thus many specialized sampling and scoring methods have been developed for this purpose over the last two decades, which perform very well for „classical“ drug design applications, namely predicting the binding of non-covalently bound small molecule-based ligands to predominantly rigid target proteins. Nevertheless, these approaches experience considerable limitations for the prediction of e.g. covalently bound and/or very flexible ligands like peptides or macrocycles as well as for flexible and/or metal containing binding sites. We develop new and optimize existing approaches for these types of binding events. Thereby our focus lies on the improvement of classical docking approaches by combination with biophysical sampling and QM/MM methods and the design of multiscale pipelines for an efficient, yet still accurate prediction of the ligand's binding characteristics. In this presentation we will discuss the advantages and limitations of such integrated approaches, present examples of successful application studies, and point to possible future application areas [1-8].

- [1] I. Antes, *Proteins*, **78**, 1084 (2010).
- [2] M. Zachmann, G. Mathias, and I. Antes, *Chem.Phys.Phys.Chem.* **16**, 1739 (2015).
- [3] M. Schneider, M. Rosam, M. Glaser, A. Patronov, H. Shah, K.C., Back, M.A. Daake, J. Buchner, I. Antes, *Proteins* **84**, 1390 (2016)..
- [4] A. Marion, J. Gora, O. Kracker, T. Frohr, R. Latajka, R., Sewald N., and I. Antes, *J. Chem. Inf. Model.* **58**, 90 (2018).
- [5] E.R. Duell, M. Glaser, C. Le Chapelain, I. Antes, M. Groll, and E.M. Huber, *ACS Chem. Biol.* **11**, 1082 (2016).
- [6] F.A. Mandl, V.C. Kirsch, I. Ugur, E. Kunold, J. Vomacka, C. Fetzer, S. Schneider, K. Richter, T.M. Fuchs, I. Antes, and S.A. Sieber, *Angew. Chem. Int. Ed. Engl.* **55**, 14852 (2016).
- [7] A. Marion, M. Groll, D.H. Scharf, K. Scherlach, M. Glaser, H. Sievers, M. Schuster, C. Hertweck, A.A. Brakhage, I. Antes, and E.M. Huber, *ACS Chem. Biol.* **12**, 1874 (2017).
- [8] I. Ugur, M. Schatte, A. Marion, M. Glaser, M. Boenitz-Dulat, and I. Antes, *PLoS One* **13**, e0205057 (2018).

## **Hybrid organic magnetic metal interfaces**

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The density functional theory provides a framework with predictive power that can be used to describe organic-metal hybrid systems in a realistic manner. In this respect, ab initio studies elucidate how the subtle interplay between the electrostatic, the weak van der Waals and the strong chemical interactions determine the geometric, electronic and magnetic structure of hybrid organic-metal interfaces. More precisely, the interaction between the  $\pi$ -like electronic cloud of organic materials with the magnetic states of a metal influences the (i) spin-polarization, (ii) magnetic exchange coupling, (iii) magnetic moments and (iv) their orientation at the hybrid interfaces. In this talk I will briefly summarize how first-principles calculations (i) provide the basic insights needed to interpret surface-science experiments and (ii) are a key tool to design novel materials with tailored properties that can be integrated in carbon-based spintronic devices.

- [1] N. Atodiresei, J. Brede, P. Lazi, V. Caciuc, G. Hoffmann, R. Wiesendanger, S. Blügel, *Phys. Rev. Lett.* **105**, 066601 (2010).
- [2] M. Callsen, V. Caciuc, N. Kiselev, N. Atodiresei, S. Blügel, *Phys. Rev. Lett.* **111**, 106805 (2013).
- [3] K. V. Raman, A. M. Kamerbeek, A. Mukherjee, N. Atodiresei, T. Sen, P. Lazić, V. Caciuc, R. Michel, D. Stalke, S. K. Mandal, S. Blügel, M. Münzenberg, J. S. Moodera, *Nature* **493**, 509 (2013).
- [4] J. Brede, N. Atodiresei, V. Caciuc, M. Bazarnik, A. Al-Zubi, S. Blügel, R. Wiesendanger, *Nature Nanotechnology* **9**, 1018 (2014).
- [5] F. Huttmann, A. J. Martínez-Galera, V. Caciuc, N. Atodiresei, S. Schumacher, S. Standop, I. Hamada, T. O. Wehling, S. Blügel, and T. Michely, *Phys. Rev. Lett.* **115**, 236101 (2015).
- [6] B. Warner, F. El Hallak, N. Atodiresei, Ph. Seibt, H. Prüser, V. Caciuc, M. Waters, A. J. Fisher, S. Blügel, J. van Slageren, C. F. Hirjibehedin, *Nature Communications* **7**, 12785 (2016).
- [7] M. Passens, V. Caciuc, N. Atodiresei, M. Feuerbacher, M. Moors, R. E. Dunin-Borkowski, S. Blügel, R. Waser and S. Karthäuser, *Nature Communications* **8**, 15367 (2017).

## **Multiscale modeling of carbon-carbon supercapacitors by a lattice model method**

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Supercapacitors store energy at the interface between an electrode and an electrolyte by ion adsorption. While these devices deliver a considerable amount of power, the energy density that they can store remains low for commercial applications. Improving the performances of such systems requires a thorough characterisation of the electrode/electrolyte interface. To this end, molecular simulations are a powerful tool, as they allow for a local description of the systems. However, the difference in scale between systems considered in these approaches and samples considered for experiments makes the comparison between theoretical and experimental data, and therefore gain of insights, a less straightforward step.

In this context, we adapted a lattice model approach [1] to the study of porous carbon supercapacitors to take advantage of its multiscale nature. The scheme we developed uses data extracted from molecular simulations, such as molecular dynamics or density functional theory, to investigate structural and dynamical properties of systems modelled at the carbon particle scale. Since the model grants access to these properties at a much lower computational cost than molecular simulations, it allowed us to consider a range of systems with different physico-chemical properties both on the electrode and the electrolyte side. In this work, we considered 10 different carbons, with well defined pore sizes, and 4 electrolytes.

We show that the storage mechanism involves ionic exchange, i.e. simultaneous counter-ion adsorption and co-ion desorption. The number of ions actually involved in the adsorption/desorption events depends on the porous material and on the electrolyte. The ionic exchange is more pronounced in small micropores compared to larger pores. We also examine the solvent effect and show that the presence of acetonitrile in the electrolytes enhances the ionic exchange. The diffusion coefficients calculated are in agreement with the ionic exchange results. In particular, in our model, carbons with smaller pore sizes favor fast diffusion, with solvation enhancing the diffusivity even more. Moreover, capacitive properties show a strong dependence on the nature of the electrode and electrolyte. In general, higher ionic charge storage occurs in systems containing neat ionic liquids. Carbons with small pores deliver larger capacitance when in contact with pure ionic liquids, while organic electrolytes give better performances when combined with carbons containing larger pores.

[1] C. Merlet et al., *J. Chem. Phys.*, **142**, 094701 (2015).

## **Plane waves for molecular dynamics - potentials and perspectives**

**Martin P. Bircher**, Ursula Rothlisberger

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Over the last decades, sampling efficiency of density functional theory based first-principles molecular dynamics (FPMD) simulations has increased considerably. With the sampling error itself decreasing, the availability of more accurate, but cost-effective exchange-correlation (xc) functionals has become of central importance to the field. FPMD has substantially profited from developments for static first principles calculations, which implies that many of the most promising xc functionals available to date have been tested (or even designed) with atomcentred, Gaussian basis sets in mind. Historically, however, plane waves have been the method of choice for many FPMD implementations. Full transferability between results obtained in an atom-centred picture such as the one represented by Gaussian functions and a delocalised basis set like plane waves is often assumed, which is of fundamental importance regarding the expected accuracy. Here, we will use the example of several xc potentials to further investigate on this core assumption. We first examine the plane-wave implementation of the Coulomb-attenuation method (CAM), a common choice for the description of charge-transfer excitation energies in linear-response time-dependent DFT. We demonstrate that CAM exhibits excellent basis-set transferability but stress at the same time that commonly employed Gaussian basis sets will not necessarily provide converged spectra, and that converged excitations can be most straightforwardly obtained from plane waves [1]. We then move on to show that transferability cannot always be taken for granted. Using the popular family of Minnesota functionals and several reaction enthalpy examples we show that in plane waves, converged energetics require an increase of the ratio between wavefunction and density cutoff. Only once the integration mesh is sufficiently dense, converged values can be easily obtained. We compare these values to their counterparts obtained in atom-centred Gaussian and Slater bases and show that, depending on the functional, substantial differences are observed - even between different Gaussian basis sets which are in principle expected to be of comparable accuracy [2]. Plane waves

therefore provide a valuable tool for obtaining reference values at the basis set limit, in particular for sensitive xc approximations. Given the possibility of obtaining fully converged basis-set limit results, it is desirable to speed up the evaluation of the ubiquitous exact exchange integrals which is substantially more costly than in a Gaussian basis. We close our exposé by demonstrating that, for isolated systems, speedups of one order of magnitude are possible by exploiting the uniform coordinate-scaling of the exact exchange functional [3], making larger scale exact exchange calculations on conventional infrastructure accessible for system sizes that have previously been prohibitive, offering new perspectives for plane-wave FPMD - sampling longer time scales with more accurate xc approximations.

- [1] M.P. Bircher and U. Rothlisberger : *J. Chem. Theory Comput.* **14**, 6, 3184 (2018)-  
<https://doi.org/10.1021/acs.jctc.8b00069>.
- [2] M. P. Bircher, P. López-Tarifa and U. Rothlisberger : *J. Chem. Theory Comput.* **15**, 557 (2019)-  
<https://doi.org/10.1021/acs.jctc.8b00897>.
- [3] M. P. Bircher and U. Rothlisberger : *J. Phys. Chem. Lett.* **9**, 14, 3886 (2018)-  
<https://doi.org/10.1021/acs.jpcllett.8b01620>

### ***Complete mapping of interfacial tension and kinetic growth coefficients of bcc and fcc metals by molecular dynamic simulations of large systems using graphic processing units***

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The shape and growth rate of crystals in metastable liquids are mainly determined by the magnitude and anisotropy of the crystal-liquid interfacial tension ( $\gamma$ ) and of the kinetic growth coefficient ( $\mu$ ). A complete mapping of both quantities of Ni (FCC) and Ti (BCC) is performed by simulations of inhomogeneous crystal-liquid systems for different crystal orientations in large systems. The interfacial tension is obtained from the analysis of capillary waves of coexisting systems while the kinetic growth coefficient from the propagation velocity of a crystal in a liquid. Using expansions of  $\gamma$  and  $\mu$  in cubic harmonics it is found that the anisotropy in bcc systems is much lower in that in fcc systems and that in both cases  $\gamma_{100} > \gamma_{110} > \gamma_{111}$  and  $\mu_{100} > \mu_{110} > \mu_{111}$ . The choice of a thermostat and its coupling strength is crucial since the crystal growth velocity of a crystal depends on it. These conclusions are based on simulations of about  $10^6$  atoms on time scales from 1 to 10 ns with many body embedded atom method (EAM) potentials using parallel codes implemented in CUDA language for graphic processing units (GPU).

### ***Designing transport properties of graphene nanoribbon junctions***

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The bottom-up chemical self-assembly approach allows synthesizing graphene nanoribbons (GNR) of well-defined width and chirality, their junctions, and can potentially be used for producing complex all-graphene electrical circuits. The presence of GNR junctions in the interconnects between individual electrical components of such circuits is unavoidable. In our work, we address the electronic transport properties of 60 and 120 degrees angled junctions that connect two GNR leads of identical width and chirality. We perform an exhaustive exploration of possible configurations of angled GNR junctions in order to identify the structures characterized by the highest ballistic transmission. Our results are rationalized in terms of simple structural descriptors of the studied angled junctions.

### ***Quantum-mechanical modeling of organic-inorganic hybrid materials as heterogeneous catalysts for pharma industry***

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Developing efficient heterogeneous catalysts for pharmaceutical industrial processes in the C-C coupling reactions is a current major challenge for material scientists, organic chemists, industrial engineers and other researchers. Recently, multifunctional organic-inorganic hybrid materials have become an attractive alternative class of catalysts, for applications in cascade, asymmetric, multi-step reactions. The idea of



designing hybrid materials is originated by the observation of the success of nature developing enzymes in living organisms. Indeed, selectivity is a crucial factor for the survival of the species. Organic-inorganic hybrid materials combine the advantages of inorganic solids (high mechanical, thermal, and structural stability) and organic molecules or macromolecules (flexibility and functionality), maximizing the chemical efficiency. Beside the different synthetic strategies, the covalent interactions among organic and inorganic units to synthesize class II hybrids allow to obtain more robust and stable functional solids. Nevertheless, the family of Class II hybrid materials could suffer of low conversions and low selectivity because of a non-regular distribution of the different active centers. In this context, also thanks to the recent evolutions in High Performance Computing (HPC) architectures, theoretical methods can be successfully applied to provide structural information and interface properties at an atomistic level to both guide and interpret experimental synthesis and measurements. In this contribution, periodic and cluster models of silica-based frameworks, together with realistic models of hydroxylated amorphous silica surfaces, functionalized with (metal)organic catalysts, will be reported as reference for the synthesis and characterization of innovative porous organic-inorganic materials, with several active sites. The advantages provided by the joint use of experimental and computational techniques will be also highlighted.

### ***Discovering surface designs to nucleate metastable polymorphs***

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Exploiting desired properties from materials has great importance to many emerging technologies. Changing a materials polymorph is arguably the most fundamental change one can make to its utility. This affects an array of properties ranging from mechanical to the band gap of semiconductors. For instance, polymorphism's effect on physiochemical properties is vital to the pharmaceutical industry. Drugs delivered in the wrong polymorph can have catastrophic consequences for the patient, as occurred with the high-profile case of ritonavir[1]. However, obtaining desired polymorphs can prove exceptionally difficult when considering metastable states. This difficulty is amplified when considering materials that show stacking disorder, where consecutive layers of different polymorphs can stack seamlessly on top of one another. I will present a methodology for discovering novel surface designs which act as templates for the controlled growth and nucleation of a metastable polymorph for materials that exhibit stacking disorder. Taking epitaxy as the inspiration we first take a pristine copy of the polymorph as a template for growth and simulate its nucleation. Simplifying and extracting contact layers leads to the discovery of novel designs which nucleate the metastable polymorph. These designs can then be matched to real surfaces to identify candidate systems that can be used in the lab. We illustrate this method on cubic ice (Ic), the metastable polymorph of Ice I under ambient conditions. A macroscopic crystal of Ic has never been achieved, and it has important possible applications in cryopreservation and weather science [2–5]. Many different hydroxyl group surfaces were discovered to nucleate Ic, and we present interesting novel designs which have no symmetry to the Ic lattice. Our hope is that this work will help to achieve this elusive polymorph in experiment and inspire work to achieve other polymorphs in ice and beyond.

[1] J. Bauer, S. Spanton, R. Henry, J. Quick, W. Dziki, W. Porter, J. Morris, *Pharm. Res.* **18**, 859 (2001).

[2] J.E. Shilling, M.A. Tolbert, O.B. Toon, E.J. Jensen, B.J. Murray, A.K. Bertram, *Geophys. Res. Lett.* **33**, 1 (2006).

[3] J. Goodman, O.B. Toon, R.F. Pueschel, K.G. Snetsinger, S.J. Verma, *Geophys. Res. Atmos.* **94**, 16449 (1989).

[4] J. Dubochet, M. Adrian, J.-J. Chang, J.-C. Homo, J. Lepault, A.W. McDowell, P. Schultz, *Rev. Biophys.* **21**, 129 (1988).

[5] P. Mehl, P. Boutron, *Cryobiology* **25**, 44 (1988).

### ***Free energy landscapes at the interface between materials and the biological world***

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Interactions at the abiotic/biotic interface have become of eminent interest in fields ranging from basic research, pharmaceutical formulation development, up to industrial product design, leaving several open challenges. Particularly, the interaction of silica (SiO<sub>2</sub>) with biological systems is complex and contradictory. For example, on the one hand, silica is at the basis of several biomineralization processes (e.g. in sponges) while, on the other hand, silica nanoparticles and dust may lead to silicosis and, at the cellular level, hemolysis. This behavior could be related to similar mechanisms of molecular recognition

between the cellular membranes and the surface of the SiO<sub>2</sub> particles. Furthermore, adsorption of enzymes on silica surfaces may lead to conformational changes that reduce their catalytic conversion activity and are thus detrimental to the efficiency of biotechnology or biosensing applications of this material. Molecular dynamics (MD) simulations are a valuable tool in investigating these interactions. However, despite the increase in available computational resources, MD still suffers a debilitating timescale problem that greatly reduces the number of phenomena that can be investigated. Numerous enhanced sampling methods have been introduced to alleviate this problem, such as Metadynamics (MetaD) and Replica Exchange with Solute Tempering (REST). We will here present examples of their applications, to shed light on the interactions between biomolecules and inorganic materials. Particularly, recent results will be shown regarding the investigation of silica-induced protein conformational changes[1] and the effects of silica nanoparticles of various size and features on membrane models of different composition[2].

[1] N. Hildebrand, M. Michaelis, N. Wurzler, Z. Li, J. D. Hirst, A. Micsonai, J. Kardos, A. Gil-Ley, G. Bussi, S. Köppen, M. Delle Piane, L. Colombi Ciacchi, *ACS Biomater. Sci. Eng.* **4**, 4036 (2018).

[2] M. Delle Piane, S. Potthoff, J. C. Brinker, L. Colombi Ciacchi, *J. Phys. Chem. C.* **122**, 21330 (2018).

### **Efficient equilibration of complex particulate systems**

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Particle simulations are a standard tool to study the phase behavior of fluids and solids. Traditional methods evolve the system deterministically by solving Newton's equation of motion or statistically with Monte Carlo. But both simulation methods can be trapped in long-lived metastable states and often do not reach the time scales accessible in experiment. Examples are the aging of glasses and crystallization processes. Here we present advanced methods to speed up the simulation of complex particulate systems and discuss structure formation phenomena that can be investigated with them. Newtonian event-chain Monte Carlo is a collective move simulation method that updates particles along meandering chains of collision events [1]. We apply Newtonian event chains to polyhedral particles that form hierarchical networks. Another system of interest are disperse particle mixtures that are natural outcomes of syntheses (colloids, nanoparticles) or can develop dynamically through exchange of mass (micelles) or charge (atoms). In another approach to speed up equilibration, we combine molecular dynamics with particle swap moves or particle resize moves. Such simulations efficiently equilibrate even polydisperse systems of spheres [2] and allow high-precision free energy calculations of systems of colloids confined in emulsion droplets [3]. Taken together, our results presented here reveal the bottlenecks of relaxation processes in particulate systems and the role of collective moves to efficiently update their geometric arrangement.

[1] M. Klement, M. Engel, *J. Chem. Phys.* **150**, 174108 (2019).

[2] P.K. Bommineni, N.R. Varela-Rosales, M. Klement, M. Engel, *Phys. Rev. Lett.* **122**, 128005 (2019).

[3] J. Wang, C.F. Mbah, T. Przybilla, B.A. Zubiri, E. Spiecker, M. Engel, N. Vogel, *Nature Commun.* **9**, 5259 (2018).

### **Band edge alignment at NiO/H<sub>2</sub>O and Ni<sub>2</sub>P/H<sub>2</sub>O interfaces**

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The search for semiconductor co-catalyst requires the calculation of the alignment of valence and conduction bands of the co-catalyst with respect to the redox levels of liquid water[1]. In this context, we analyze two transition metal-based co-catalyst, namely NiO and Ni<sub>2</sub>P, which have been proven to exhibit high hydrogen evolution rates when used in conjunction with metal-organic frameworks (MOFs), which act as photo-catalysts[2]. We perform hybrid-functional calculations for computing the band structure of the two semiconductors. Both molecular and dissociative models of adsorbed water at the semiconductor surfaces are considered. Finally, molecular dynamics simulations of the NiO/H<sub>2</sub>O and Ni<sub>2</sub>P/H<sub>2</sub>O interfaces are used to determine the band edge alignment.

[1] Z. Guo, F. Ambrosio, W. Chen, P. Gono, and A. Pasquarello, *Chem. Mater.* **30**, 94 (2017).

[2] S. Kampouri, T. N. Nguyen, M. Spodaryk, R. G. Palgrave, A. Züttel, B. Smit, and K. C. Stylianou, *Adv. Func. Mater.* **28**, 1806368 (2018).

## **Predicting the bilayer membrane permeability of large drugs**

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We have developed a semi-empirical simulation method that can predict the permeability of large drugs (up to MW 1500) through lipid membranes. Drug discovery is currently addressing molecular compounds that are larger than could be anticipated a few decades ago, sometimes summarized as beyond-the-rule-of-five (bRO5) compounds. One may think of protein-protein interaction inhibitors, kinase inhibitors, and all biologicals. Age-old statistical methods to predict development qualifiers such as solubility and membrane permeability, date from the times that pre-date the new bRO5 chemistries. In the classical Overton permeation model, the core of a membrane is regarded as a kind of liquid oil, which has led to many classical permeation models that base correlations on some partition coefficient descriptors (LogP, LogK(hydrocarbon), etc. We find that the classical model works fine for the small molecules of the past, but for the larger molecules bRO5 of today's interest (MW > 500), the Overton model is completely wrong: the actual permeability can be orders of magnitude (> 5 orders!) lower than predicted by the standard descriptors based on liquids phases. Our most dramatic result is that for these large molecules, the elasticity or packing constraint in the lipid membrane core outweighs the traditional liquid partition effect. To put it in simple terms: large molecules have to wiggle in between tightly packed lipid tails and is the more difficult, the bigger the drug. Our modeling results are confirmed by comparing with experimental results from Walter and Gutknecht[1] (1980's MW < 100), Xiang and Anderson[2-3] (1990-2010, MW < 300), Kraemer group[4-5] (2010-present, all drugs, MW 200-800) and the Lokey group[6] (2015-present, modified peptides MW up to 1300). The physics-based modeling we propose rests on two innovations: (a) the so-called coarse-grained paradigm, that lumps groups of atoms into small fragments, and (b) a semi-empirical method to calculate molecular charges. Our algorithms[7] permit the complete calculation of permeability for a large drug-like molecule in one go in a mere few minutes on a single core of an ordinary desktop computer. The algorithm includes automated ways for calculation of charge distribution, the cutting of molecules into pieces and, the calculation of thermodynamics and diffusion through thermodynamic integration.

[1] A. Walter, J. Gutknecht, *J. Membrane Biology* **90**, 207 (1986).

[2] T.X. Xiang, B.D. Anderson, *J. Membrane Biology* **140**, 111 (1994).

[3] Y.C. Cao, T.X. Xiang, B.D. Anderson, *Molecular Pharmaceutics* **5**, 371 (2008).

[4] K.F. Hermann, C.S. Neuhaus, V. Micalef, B. Wagner, M. Hatibovic, H.E. Aschmann, F. Paech, R. Alvarez-Sanchez, S.D. Krämer, S. Belli, *European Journal of Pharmaceutical Sciences* **104**, 150 (2017).

[5] S.D. Krämer, H.E. Aschmann, M. Hatibovic, K.F. Hermann, C.S. Neuhaus, C. Brunner, S. Belli, *Advanced Drug Delivery Reviews* **101**, 62 (2016).

[6] C.R. Pye, W.M. Hewitt, J. Schwochert, T.D. Haddad, C.E. Townsend, L. Etienne, Y.T. Lao, *J. Med. Chem.* **60**, 1665 (2017).

[7] J.G.E.M. Fraaije, J. Van Male, P. Becherer, R. Serral Gracià, *J. Chem. Inf. Model.* **56**, 2361 (2016).

## **Symmetric spin initialization: a new powerful tool for computational materials discovery**

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We present a new method that has been recently implemented in the evolutionary algorithm USPEX for the initialization of magnetic moments in crystals. The method is based on an empirical observation: the majority of the antiferromagnetic materials known today has a ground state in which the different atomic spin states break symmetry only up to a subgroup of index 2 with respect to the parent space group of the paramagnetic phase. Therefore, with our method, magnetic structures are generated in USPEX by imposing this symmetry criterion. In addition, a new calculation regime has been implemented in USPEX for the search of the ground magnetic state with fixed geometry. Two tests of our method have been performed, respectively for the optimization of structural and magnetic degrees of freedom and for the optimization of only magnetic degrees of freedom in the newly implemented calculation regime. The first test shows an improved sampling of the free energy surface of Fe<sub>12</sub>O<sub>18</sub>, while the second test correctly reproduces the ground state of hematite at the first generation. The problem of computing the U correction for DFT+U has also been addressed. Using linear response theory, a value of U = 7.6 eV has been obtained for hematite.

### ***What is the role of hydrophobic mismatch in helix dimerization?***

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Folding and packing of proteins depend on their amino acid sequence and the environment in which these processes take place. For membrane proteins, it is the lipid bilayer that determines the type and strength of the interactions that will define the mature tertiary and/or quaternary structure. Here, we explore how the hydrophobic mismatch (the difference between the hydrophobic span of a transmembrane protein region and the hydrophobic thickness of the lipid membrane around the protein) influences the transmembrane helix dimerization and packing in cellular environment. Using a ToxRED assay in *Escherichia coli* and a Bimolecular Fluorescent Complementation approach in human-derived cells, complemented by atomistic molecular dynamics simulations, we analysed the dimerization of Glycophorin A derived transmembrane segments. We showed that biological membranes can accommodate transmembrane homo-dimers with a wide range of hydrophobic lengths. Interestingly, the hydrophobic mismatch was found to be considerably weaker than previously observed in model membranes, or under in vitro conditions, indicating the presence of robust compensatory mechanisms able to alleviate membrane stress in biological membranes. Atomistic simulations revealed that these mechanisms involve primarily dimer tilting and local membrane thickness perturbations. Moreover, cells also tolerate the hetero-dimers with large length disparity between their monomers, using the same compensatory mechanisms as those found in homo-dimers. However, large differences between transmembrane helices length hinder the monomer/dimer equilibrium, confirming that, the hydrophobic mismatch has, nonetheless, biologically relevant effects on helix packing in vivo.

### ***Molecular model database and semantic technology development for materials modelling platforms and workflows***

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Where databases and platforms using different data structures and file formats interoperate, or where data from various sources are combined, agreement on semantics becomes a necessity. The openly accessible molecular model database (MolMod DB) of the Boltzmann-Zuse Society for Computational Molecular Engineering contains materials relations (intermolecular pair potentials) for over 150 fluids, meant for molecular modelling and simulation with molecular dynamics (MD) and Monte Carlo (MC) solvers [1]. The molecular models in the database have been published in about 30 articles over the past 20 years, which are associated with the respective entries and can be followed on the MolMod DB web front end. The database provides a wide range of search functionalities, e.g., for substances (names and CAS numbers) and model classes.

Semantic assets are provided to ensure interoperability with a variety of platforms, on the basis of the Review of Materials Modelling [2] conducted by the European Materials Modelling Council (EMMC). Input files for several common environments can be downloaded via the web front end, including the file formats used by the molecular simulation codes ms2 [3], GROMACS [4], LAMMPS [5], and ls1 mardyn [6]. The standardization efforts guided by the EMMC will create an opportunity to integrate modelling algorithms and tools with repositories, open translation environments, and virtual marketplaces. Accordingly, the MolMod DB is intended to be interoperable with infrastructure associated with the EMMC, including the Virtual Materials Marketplace (VIMMP); Acknowledgment: This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 760907.

[1] S. Stephan, M. Horsch, J. Vrabec, H. Hasse, H., *Molecular Simulation* **45**, 806 (2019).

[2] A. F. De Baas, *What Makes a Material Function?* EU Publications Office, Luxembourg (2017).

[3] G. Rutkai, A. Köster, G., Guevara Carrión, T. Janzen, M. Schappals, C.W. Glass, M. Bernreuther, A. Wafai, S. Stephan, M. Kohns, S. Reiser, S. Deublein, M. Horsch, H. Hasse, J. Vrabec, J., *Computer Physics Communications* **221**, 343 (2017).

- [4] M. Abraham, T. Murtola, R. Schulz, P. Szilárd, J. Smith, B. Hess, E. Lindahl, E., *SoftwareX* **1-2**, 19 (2015).  
 [5] S. Plimpton, *J. Comput. Phys.* **117**, 1 (1995).  
 [6] C. Niethammer, S. Becker, M. Bernreuther, M. Buchholz, W. Eckhardt, A. Heinecke, S. Werth, H.-J. Bungartz, C.W. Glass, H. Hasse, J. Vrabec, M. Horsch, M, *J. Chem. Th. and Comput.* **10**, 4455 (2014).

### **Study of pseudo-phase behaviour of a single flexible-semiflexible multi-block copolymer chain by means of stochastic approximation Monte Carlo algorithm**

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The combination of flexibility and semiflexibility in a single molecule is a powerful design principle both in nature and in materials science. We present our results on the conformational behavior of a single SF-multiblock-copolymer chain (of length  $N=64$ ), consisting of equal amounts of flexible (F) and semiflexible (S) blocks (of length  $b=4, 8, 16, 32$ ) with the same or different affinity to an implicit solvent [1-5]. We have used a continuum bead-spring model and Stochastic Approximation Monte Carlo (SAMC) algorithm which has been established as a mathematically founded powerful flat-histogram Monte Carlo method, used to determine the density of states,  $g(U)$ , which is related to the microcanonical entropy. We have considered a manifold of macrostates defined by two terms in the conformational energy - intermonomer interaction energy and stiffness energy - and accumulated two-dimensional density of states functions  $g(U_1, U_2)$ . In an analysis in the canonical ensemble, we calculated the heat capacity and determined its maxima and the most probable morphologies in different regions of the state diagrams (pseudo-phase diagrams). These are rich in various, non-trivial morphologies, which are formed without any specific interactions, and depend on the block length and the type of solvent selectivity (preferring S or F blocks, respectively). These pseudo-phase diagrams display multiple nematic pseudo-phases (structures, morphologies) in the collapsed state, characterized through a demixing of the blocks of different stiffness (even in a non-selective solvent) and orientational ordering of the stiff blocks. We observe dumbbell-like morphologies, lamellar phases, and for the larger block lengths also Saturn-like structures with a core of flexible segments and the stiff segments forming a ring around the core. We have also implemented microcanonical analysis in the "conformational" microcanonical (NVU, where U is the potential energy) and in the true microcanonical (NVE, where E is the total energy) ensembles with the aim to reveal and classify pseudo-phase transitions, occurring under the change of temperature. We show how taking the kinetic energy into account alters the predictions from the analysis. The funding from German Science Foundation (DFG grant PA 473/18-1) and Russian Foundation for Basic Research (RFBR grant 19-53-12006) is acknowledged.

- [1] S.V. Zablotskiy, J.A. Martemyanova, V.A. Ivanov, W. Paul, *J. Chem. Phys.* **144**, 244903 (2016).  
 [2] S.V. Zablotskiy, V.A. Ivanov, W. Paul, *Phys. Rev. E* **93**, 063303 (2016).  
 [3] S.V. Zablotskiy, J.A. Martemyanova, V.A. Ivanov, W. Paul, *Polym. Sci. Ser. A* **58**, 899 (2016).  
 [4] T. Shakirov, S.V. Zablotskiy, A. Böker, V.A. Ivanov, W. Paul, *Eur. Phys. J. Spec. Top.* **226**, 705 (2017).  
 [5] D. Maltseva, S. Zablotskiy, J. Martemyanova, V. Ivanov, T. Shakirov, W. Paul, *Polymers* **11**, 757 (2019).

### **Spontaneous binding of (anti)folate(s) to a membrane-anchored receptor witnessed by MD simulations**

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Observing spontaneous binding of bioactive ligands to their target receptors may provide valuable clues towards the recognition mechanism and, hence, enable better understanding and design of biomimetic or biocompatible systems. MD simulations of such events usually require either enhanced sampling techniques or generation of a prohibitively large number of independent trajectories. In the current study, enforced binding of folate to its membrane-transporting biomolecule - the folate receptor-alpha, is registered at the time scale of several hundred nanoseconds by equilibrium MD. It is suggested that the process is effectively triggered at such a short time scale by representing as fully as possible the natural molecular environment of the ligand-receptor pair. The simulations also capture the specificity of

interaction of several experimentally available derivatives of folate. The differences in the binding patterns are attributed to the chemical and structural variations of the ligands. Acknowledgments:

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### ***Conformational changes of NorA Multidrug efflux pump transporter in interaction with diverse drugs***

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Efflux is a key mediator of antimicrobial resistance and understanding the molecular level interaction of antibiotics with efflux pumps in bacteria, can provide useful information to design new antibiotics that can overcome efflux. Advanced computational tools offer a very powerful approach for understanding the conformational changes of the drug efflux cycle and their related interactions with the critical residues within the efflux transporter's channel. We carried out 1  $\mu$ S molecular dynamics simulation (MD) of NorA transporter from *Pseudomonas aeruginosa* in interaction with diverse compounds as substrate and inhibitors in the presence of a membrane to elucidate the molecular mechanism of the drug efflux cycle. Methods MD simulations started from the docked structures, obtained from blind docking by AutoDock SMINA, was followed by flexible docking using the GOLD program. The MD simulations were carried out in AMBER 16.0 using the AMBER forcefield. In this study, relevant residues were protonated in diverse access/binding/extrusion monomers of the NorA transporter to study the conformational transition during the long MD simulations in free and bound forms of NorA. Results In this study, we were able to extend the understanding of the NorA efflux pump to show significant differences between the binding and free forms of NorA. In molecular dynamics simulations, the consequence of substrate interactions could also be seen regarding notably different conformational changes in the structure of the transporter compared with the free form and the pump in complex with inhibitors. MD simulation analysis revealed the critical residues within the binding pocket that guide the substrate through the binding site. This study provides a molecular basis for understanding the mechanism of efflux of an antibiotic by the NorA transporter. Conclusion The observed conformational changes of the transporter - adopted by the substrate-bound form - are different from the observed corresponding results for the inhibitor-bound form of the transporter. The results could help to broaden our knowledge and shed some more light on the mechanism of tripartite efflux pumps.

### ***Numerically accurate GW approach to electronic band structure of weakly and strongly correlated materials***

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Many-body perturbation theory in the GW approximation is currently regarded as the most accurate and robust first-principles approach to electronic band structure of weakly correlated insulating materials. Recent systematic studies of ZnO by several groups clearly indicate the importance of numerical accuracy in the practical implementation of the GW methods. In this talk I will address the challenges for numerically accurate GW calculation based on our recent systematic investigation of the effects of including high-energy local orbitals (HLOs) in the linearized augmented plane waves (LAPW)-based GW calculations for both weakly and strongly correlated materials [1-3]. It is shown that both the accuracy of unoccupied states and the completeness in the summation of states are crucial for numerically accurate GW calculations. In general, using LAPW+HLOs basis can significantly improve the performances of the semi-local density functional approximation based GW0 approach [2]. We have further investigated other systems with electronic configurations that are significantly different from those of common sp-semiconductors, including VII-IB compounds (MX with M=Cu, Ag and X=Cl, Br and I) and strongly correlated d- or f-electron oxides (NiO, Ce2O3 and UO2) [3]. We found that the consideration of HLOs in the GW based on density-functional theory plus the Hubbard U-correction (GW0@DFT+U) approach can significantly improve the description of electronic band structure of those d- and f-electron systems.

[1] H. Jiang, R. I. Gomez-Abal et al. *Computer Phys. Commun.* **184**, 348(2013).

[2] H. Jiang and P. Blaha, *Phys. Rev. B* **93**, 115203(2016).

[3] H. Jiang, *Phys. Rev. B* **97**, 245132(2018).

### **Computational study of Y NMR shielding in intermetallic Yttrium compounds**

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Density functional theory (DFT) calculations of the magnetic shielding for solid state nuclear magnetic resonance (NMR) provide an important contribution for the understanding of experimentally observed signals. In this work, we present calculations of the Y NMR shielding in intermetallic compounds. (YMg, YT, YTX, YT<sub>2</sub>X, YT<sub>2</sub>X<sub>2</sub>, Y<sub>2</sub>TB<sub>6</sub> and Y<sub>2</sub>TSi<sub>3</sub> where T represents various transition metals and X refers to group IV elements C, Si, Ge, Sn, Pb). The total shielding sigma of this selection varies by about 2500 ppm and correlates very well with the experimentally observed shifts except for YMg and YZn. These two simple compounds have a spike in the DOS at Energy Fermi and a corresponding huge spin susceptibility which leads to the disagreement. It could be a problem of DFT (neglect of spin fluctuations), but we would interpret the discrepancies as caused by disorder which could be present in the experimental samples, because disorder removes the spike in the DOS. The diamagnetic contribution sigma-o (chemical shift) is by no means constant as often assumed when interpreting experimental metallic shifts and varies up to 1500~ppm, but still the dominating term is the spin contact term sigma-c. Although all compounds are metals, only half of them have a paramagnetic (negative) sigma-c due to the reoccupation of the valence Y-5s electrons, while for others the large induced Y-4d magnetic moment induces a diamagnetic core polarization. In most of our cases, the spin dipolar contribution sigma-sd is fairly small with |sigma-sd| less than 100 ppm, and often even much smaller except in a few very asymmetric compounds like YCo<sub>2</sub>Si<sub>2</sub> and YRu<sub>2</sub>Si<sub>2</sub> sigma-sd approx 320~ppm).

### **Electronic states in electron-doped rare-earth nickelates from first principles**

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Correlation effects in transition metal based materials give rise to many interesting and exotic properties. The rare-earth nickelates, with a rich composition-phase diagram, are no exception. Doping rare-earth nickelates can lead to electron localization, introducing defect states that are unlike typical shallow or deep donor states familiar in conventional semiconductors. We present first-principles density-functional-theory-based calculations of rare-earth nickelates, with a focus on samarium nickelate, in which we add electrons to the material. Here, we investigate doping concentrations on the order of one electron per formula unit with the goal of changing the orbital occupation and triggering a phase transition, akin to the phase control seen with strain modulation. We carry out calculations where a uniform compensating background charge ("jellium") has been added to maintain charge neutrality when electrons are added, as well as supercell configurations with defects that electron dope the system. In particular, we explore the effects of intercalated hydrogen and lithium as well as oxygen vacancies in samarium nickelate. In comparing these calculations, we find the jellium-background calculations capture the changes to the electronic structure seen with the explicit inclusion of defect. The resulting changes to the electronic structure, intimately linked to structural changes, cannot be understood with a rigid shift of the states: the bands are reorganized and the character of the gap is fundamentally altered. This class of doping effects introduces a new knob to turn in the field of materials design.

### **Master equation with first-principles-derived rates for modeling of electronic relaxations in thin metallic films on Si(111)**

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The lifetime of excited electrons in atomically-thin metallic films is limited by both electron-electron (e-e) and electron-phonon (e-ph) scattering. Specifically, Pb films on Si(111) provide an ideal 'probing ground' since they comprise both dispersive bands with metallic character and confined, thickness-dependent quantum well states. We employ the description by a Master equation for the electronic occupation numbers to analyze the relative importance of both scattering mechanisms. The electronic and phononic

band structures, as well as the matrix elements for electron-phonon coupling within deformation potential theory were obtained from density functional calculations. The contribution of impact ionization processes to the lifetime is estimated from the imaginary part of the electronic self-energy calculated in the GW approximation. By numerically solving the Master equation for the occupations of the Pb-derived electronic states coupled to a phononic heat bath, we are able to follow the distribution of the electronic excitation energy to the various modes of Pb lattice vibrations. While e-e scattering is the dominant relaxation mechanism, we demonstrate that the e-ph scattering is highly mode-selective, with a large contribution from surface phonons. The time scales extracted from the simulations are compared to experimental data from time-resolved pump-probe experiments.

### ***Proton momentum distributions in strong hydrogen bonds in the solid state***

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Neutron Compton scattering (NCS) is a unique experimental technique made possible by the development of epithermal neutron sources, such as the ISIS source of the Rutherford Appleton Laboratory in the UK [1, 2]. Dynamic structure factors, measured in NCS, are solely determined by the nuclear momentum distribution (NMD). In the picture of purely classical nuclei, the NMD shape is determined by whole energy spectrum of the motional modes, including translational and rotational modes, followed by lattice and internal molecular vibrations. However, more and more experimental evidence has been accumulated over the years that nuclear quantum effects, such as nuclear zero point motion, delocalisation and tunnelling, determine the shapes of NMDs of lightweight isotopes such as protons and deuterons. At sufficiently low temperatures, all nuclear quantum systems are cooled down to their ground states. In this low-temperature limit, the NCS recoil peak shape for a given nucleus is proportional to the square of the absolute value of its nuclear wave function, which is dictated by the shape of the local, effective Born-Oppenheimer (BO) potential [1, 2]. Furthermore, different shapes of the BO potentials can be selected by applying Bayesian approach to fitting data obtained from an NCS experiment [3]. Such statistical tests can detect traces of self-interference of a nuclear wave function in effective BO potentials, a prerequisite of nuclear quantum tunnelling in condensed matter systems. Molecular crystals exhibiting strong hydrogen bonds seem as natural fit for the NCS technique. In this contribution, the results of recent NCS investigation of the solid solutions of equimolar water-phosphoric acid mixture and its deuterated counterpart, will be presented. The analysis of the NMDs, augmented with Bayesian inference methodology, reveals line-shape features characteristic for proton tunnelling in the water-H<sub>3</sub>PO<sub>4</sub> mixture below 160 K but shows no such features in the case of the deuterated water-D<sub>3</sub>PO<sub>4</sub> mixture. Taken together, these observations suggest the existence of the so-called tunnelling effect in the kinetics of the proton transfer below 160K, most likely involving concerted proton tunnelling along Grotthuss chains. It is the interplay between the amount of the ZPE and the height of the activation barrier for the proton transfer, which in consequence leads to a non-trivial nuclear quantum isotope effect, whereby kinetic rate constants of protons are orders of magnitude higher than those for deuterium. The presented methodology paves the way for a novel experimental screening protocol for the presence of the signatures of nuclear quantum tunnelling in condensed matter systems.

[1] C. Andreani *et al.*, *Adv. Phys.* **66**, 1 (2017).

[2] " F. Fernandez-Alonso and D. L. Price Eds., *Academic Press*, 2017

[3] M. Krzystyniak, *et al.*, *Phys. Chem. Chem. Phys.* **19**, 9064 (2017).

### ***Molecular dynamic simulations of ion adsorption in nanoporous carbons: systematic variation of ion size and pore structure***

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Carbon-carbon supercapacitors are a type of electrochemical energy storage systems, which use porous carbons as electrodes and store the charge at the electrolyte/electrode interface through ion adsorption. Examining the relationship between the system characteristics (pore size distribution and structure of the carbon electrode, ionic liquid nature, ion size, etc..) and its electrochemical properties (power density, energy density and capacitance), as well as understanding the molecular processes occurring in these supercapacitors can allow for their optimization.



In this study we characterise via molecular dynamics simulations a pure ionic liquid in contact with a porous carbon. Two types of carbon structures have been studied: ordered and disordered, each carbon being characterised by its own pore size distribution. This study allows us to understand the influence of the structural properties of the porous materials on the structural and dynamical properties of the adsorbed electrolyte. To characterize the local structure of the ionic liquid, we typically calculate ionic densities, pair distribution functions and degrees of confinement in order to get information on the adsorption behaviour of the confined ions. We also determine diffusion coefficients which give us insights into the dynamics of the ions under confinement. This study constitutes a first screening of nanoporous carbons which will be studied as electrode materials in model supercapacitors. In this second step, two model supercapacitors have been investigated. The electrodes are ordered for one of the systems and disordered for the other, and we examine the effect of the potentials applied on the same structural and dynamical properties as for neutral carbons.

### ***Defect-mediated melting of two-dimensional active matter***

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The nature of melting in two-dimensional (2D) solids has remained controversial for decades. It has been debated since the 30s when Landau, Peierls, and Mermin showed that thermal long-wavelength fluctuations destroy long-range positional order in 2D. Then, according to the Kosterlitz-Thouless-Halperin-Nelson-Young (KTHNY) theory, melting in 2D proceeds via two consecutive transitions, mediated by the creation/annihilation of topological defects. However, the melting scenario of the simplest 2D particle system, i.e. the hard-disk model, was clarified only a few years ago: as the density is increased, a first order phase transition between a liquid and hexatic occurs, followed by a Kosterlitz-Thouless transition between the hexatic and solid.

Here, we establish the 2D melting scenario of hard disks in the presence of non-equilibrium, active forces. We present a precise computational analysis of the equations of state, correlation functions and statistics of local order parameters of Active Brownian Particles (ABP), arguably the most studied active particle model system. We show how motility-induced phase separation (MIPS), occurring at high activities in systems of ABP, interferes with the equilibrium melting scenario. At small activities the hexatic-liquid coexistence region is maintained. An active hexatic exists for all activities in a density range which widens as activity is increased. At higher densities, an active solid phase emerges. At higher activities, we show that self-propulsion triggers a phase separation (MIPS) between a dilute and a dense phase, which can either be liquid, hexatic or solid. We characterize the behavior of topological defects across these transitions and establish the role played by localized defects and extended grain boundaries in the destabilization of the active solid and hexatic phases.

Overall, we discuss how self-propulsion affects the liquid and solid phases of matter and the nature of the non-equilibrium phase transitions between them.

### ***Symmetry aspects of spin filtering in molecular junctions: hybridization and quantum interference effects***

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Control and manipulation of electric current and especially its degree of spin polarization across single molecules is currently of great interest in molecular spintronics. Using state-of-the-art ab initio transport calculations, we explore one of possible strategies based on the modification of nanojunction symmetry which can be realized, for example, by a mechanical strain. Such modification can activate new molecular orbitals which were inactive before due to their orbital mismatch with electrode's conduction states. This can result in several important consequences such as: i) a significant suppression of the majority spin conductance was found in low symmetry configurations due to quantum interference effects seen as Fano-like features in electron transmission functions and ii) strongly enhanced conductance of minority

spin due to increased molecule-metal hybridization when the symmetry is lowered. We illustrate the idea on two basic molecular junctions: Ni/Benzene/Ni (perpendicular vs tilted orientations) and Ni/Si chain/Ni (zigzag vs linear chains). We believe that our results may offer new potential route for creating molecular devices with a large on/off spin polarization via quantum interference effects.

### ***Thermoelectricity-magnetism coexisting in (Mn, Fe, Co)-doped NiSi compounds: ab-initio study***

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Doping NiSi with thermoelectric (TE) elements (such as Te, Sb, Bi) didn't further improve its thermoelectricity, but inducing (Mn, Fe, Co) transition metals (TMs) not only maintain the TE properties but also gave rise to magnetism in the obtained alloys. The Seebeck coefficient of Mn-doped NiSi was obtained to be  $-50\text{V K}^{-1}$  at 750 K and a higher figure merit about 0.11 above room temperature (750 °K), while a magnetic moment of 2.70  $\mu\text{B}$  was found. We focus here on a possible coexistence of magnetism-thermoelectricity in TMs-doped NiSi systems under pressures up to 23 GPa (to maintain them crystallizing in a orthorhombic crystalline structure), via first-principles calculations. The results show that though TE properties did not further improved, magnetism and thermoelectricity could coexist Mn-based silicides.

### ***Colloids on curved surfaces: coupling of phase and location***

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In uniform three-dimensional space and flat two-dimensional space, the various states of matter can exist anywhere because of the translational and rotational invariance of the space. In two dimensions, the introduction of uniform curvature (the surface of a sphere) can strongly affect structure, phase transitions and dynamics of particles confined to the surface [1-3], but all locations are still equivalent. In contrast, on non-uniformly curved surfaces, different states of matter may have structural or thermodynamic preferences for regions of different curvature [4]. Hence, phase transitions may be accompanied by the migration of matter to a new position. We demonstrate this coupling of phase and location for colloids on toroidal and sinusoidal surfaces, showing that it is a general effect in the presence of non-uniform curvature. Furthermore, the nature of the coupling is strongly influenced by the range of the attractive interactions between the particles. As befits a celebration of CECAM, the work deploys bespoke simulation methodology in several areas: Monte Carlo sampling, constrained dynamics and global optimisation.

[1] G. Meng, J. Paulose, D. R. Nelson and V. N. Manoharan, *Science* **343**, 634 (2014).

[2] S. Paquay, H. Kusumaatmaja, D. J. Wales, R. Zandi and P. van der Schoot, *Soft Matter* **12**, 5708 (2016).

[3] J. O. Law, A. G. Wong, H. Kusumaatmaja and M. A. Miller, *Mol. Phys.* **116**, 3008 (2018).

[4] L. R. Gómez, N. A. García, V. Vitelli, J. Lorenzana and D. A. Vega, *Nature Comm.* **6**, 6856 (2015).

### ***From Markov state models to absolute binding free energies***

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Markov state models (MSMs) are increasingly used to combine information from many short molecular dynamics trajectories to characterize biomolecular dynamics. Based on the assumption that conformational dynamics are independent of the history of states, MSMs fully describe the thermodynamics and kinetics of the states. Given the rich information contained within MSMs, it would be advantageous to exploit them in binding free energy calculations. The statistical mechanics linking MSMs to binding free energy calculations is provided by implicit ligand theory (ILT) [2,5]. ILT specifies that noncovalent binding free energies between a flexible ligand and receptor may be determined through an exponential average of the binding potential of mean force (BPMF) - the binding free energy between a flexible ligand and rigid receptor. The rigid receptor conformations must be drawn from or reweighed to an apo [4] or holo [2] ensemble, as is possible in MSMs. To demonstrate this approach, we construct a

MSM of T4 lysozyme based on 4 microseconds of molecular dynamics initiated from four different crystal structures representing open, intermediate, and closed conformations of the enzyme. Representative conformations are selected based on the Jaccard distance between occupancy fingerprints [1]. BPMFs for 141 small organic molecules are computed for the representative snapshots using our program AIGDock [4]. Free energy estimates were compared to previous results in which snapshots were selected from alchemical binding free energy calculations [3].

[1] B. Xie, J.D. Clark, D.D.L. Minh, *J. Chem. Inf. Model.* 58, 1915 (2018).

[2] T.H. Nguyen, D.D.L. Minh, *J. Chem. Phys.* 148, 104114 (2018).

[3] B. Xie, T.H. Nguyen, D.D.L. Minh, *J. Chem. Theory Comput.* 13, 2930 (2017).

[4] D.D.L. Minh, *Protein-Ligand Binding Potential of Mean Force Calculations with Hamiltonian Replica Exchange on Alchemical Interaction Grids*. arXiv:1507.03703v1.

[5] D.D.L. Minh, *J. Chem. Phys.* 137, 104106 (2012).

### **Neural network-based study of point defect diffusion in lead telluride and cadmium telluride**

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We present recent results of neural network-based molecular dynamics (MD) simulations of point defect diffusion in lead telluride (PbTe) and cadmium telluride (CdTe). Those two compounds possess similar lattice constants but distinct crystal structures. The latter leads to their immiscibility, which, as shown in the experiments, exists in the crystal phase even at very high temperatures. During PbTe/CdTe multilayer crystal growth the structures undergo morphological transformations, which can be employed in controlled manufacturing of various nanostructures such as nanowires and quantum dots. The underlying microscopic mechanism of those transformations is currently unknown but it is expected that diffusion of cation interstitials and vacancies across the PbTe/CdTe interface plays an important role. The interatomic potential used in our simulations is a neural network potential trained with ab initio reference energies and forces obtained for a set of over 2000 configurations based on the PBEsol exchange-correlation functional. That approach allows to study systems with the accuracy comparable with ab initio MD but in much larger size and time scales, which makes it suitable for the analysis of bulk diffusion in solid bodies. By means of the MD simulations of point defects in PbTe and CdTe we determine their diffusion mechanism. We find that in PbTe cation interstitials diffuse mainly through exchange with lattice atoms while in CdTe they mostly hop between neighbouring interstitial sites. By measuring the diffusion coefficient at various temperatures we can extract the activation energy and compare it for different defects. Understanding the diffusive processes in PbTe and CdTe is an important step in the analysis of the exchange processes occurring across their interface.

### **Mechanical behavior analysis of metal-based nanocomposites via MD simulations: fundamentals, concepts, and underlying mechanisms**

**A. Montazeri**

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Regarding my experiences in the field of MD simulations of metal-based NCs having crystal structures within them, as can be found in my C.V., it will be an honor for me to present a talk in the valuable conference (if it is acceptable from the organizers). The proposed title and abstract is as follows: Mechanical behavior analysis of metal-based nanocomposites via MD simulations: Fundamentals, concepts, and underlying mechanisms. In the first part of the planned presentation, the influence of surface and volumetric porosities on the plastic deformation of metal-based nanocomposites (NCs) is thoroughly examined. The underlying mechanism governing this behavior is also discussed in details through microstructural characterization. This is followed by inspection of the temperature role on the plastic deformation of these NCs. Then, I will utilize MD simulation of nanoindentation process to explore the dominant factors influencing the nanotribological characteristics of metal matrices in the presence of graphene sheets. The presentation will be ended with introducing a new MD/FE hierarchical multiscale approach to provide a route for investigation of these NCs with the accuracy of MD and speed of finite element method (FEM).

### ***Shape-memory behavior analysis of polylactic acid/graphene nanocomposites: a molecular dynamics study***

**A. Montazeri**, A. Amini, K. Hasheminejad

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Thermally induced shape-memory polymers (SMPs) are able to recover their original shape with a change in the environment temperature after being deformed. In this regard, Polylactic acid (PLA) has attracted great attention due to its shape-memory behavior activated by heating. This behavior can be highly influenced by the hard segment content of the SMP. It is worth noting that some nanoparticles and nanoplatelets, act as a hard phase in the polymeric matrix leading to enhanced shape-memory behavior of polymers. To explore the matter, in the present work, we have applied an atomistic molecular dynamics simulation to analyze the effect of incorporated graphene platelets on the shape-memory behavior of the amorphous switching domains of PLA. By the end of this study, we may answer how much the glass transition temperature is changed by adding a hard segment. This is followed by thorough analysis of mechanisms governing the shape-memory behavior of PLA/graphene nanocomposites. It is found that the neat PLA models show higher shape recovery. In contrast, graphene/PLA nanocomposites pose higher shape fixity and lower recovery rate during the thermomechanical cycle. Our simulations reveal that functionalized graphene sheets have more pronounced effects on the overall shape-memory characteristics of PLA leading to its better performance as one of the most important SMPs.

### ***Impact of pathogenic mutations of the GLUT1 glucose transporter on channel dynamics using ConsDYN***

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Several pathogenic mutations are reported in the glucose transporter subfamily SLC2, which are known to cause several disorders such as the Glut1-deficiency syndrome. Understanding the link between these mutations and transporter dynamics is crucial to elucidate their role in the dysfunction of the underlying transport mechanism. Here, insight from molecular dynamic simulations is required to identify the molecular effects on the transporter function. We studied a variety of pathogenic and non-pathogenic mutations, using a newly developed coarse-grained simulation approach 'ConsDYN', which allows the sampling of both inward-open and outward-occluded states. To guarantee the sampling of large conformational changes, we only include conserved restraints of the elastic network introduced upon coarse-graining, which showed similar reference distances between the two conformational states ( $\leq 1$  Å difference). Hence, we capture the 'conserved dynamics' between both states. This approach is sufficiently sensitive to capture the effect of different mutations, and our results clearly indicate that the pathogenic mutation in GLUT1, G91D, situated at the highly conserved RGXRR motif between helices 2 and 3, has a strong impact on the channel function. Using our approach, we can explain the pathogenicity of the mutation G91D when we observe the configurations of the transmembrane-helices, suggesting that their relative position is crucial for the correct functioning of the GLUT1 protein.

### ***Design and implementation possibilities of a fast multipole method on FPGA***

**Zoltán Nagy**, András Kiss, Levente Márk Sántha, Péter Szolgay, György Csaba,

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Fast multipole methods (FMMs) grew out of the fast Fourier transform method and they are powerful mathematical techniques to efficiently calculate pairwise interactions in physical systems, e.g. in micromagnetic simulations. The precise simulation of the magnetization dynamics in ferromagnetic materials requires the precise estimate of the magnetostatic field, which describes the long range interactions between magnetic dipoles. By using a simple brute force algorithm field calculation can be computed in  $O(N^2)$  steps but using the FMM it can be carried out in  $O(N \log(N))$  steps or even  $O(N)$  in some special cases. The paper describes the implementation of the FMM on a reconfigurable computer, Field Programmable Gate Array (FPGA), in order to accelerate the computations of a micromagnetic

simulator. The possible computing sizes on FPGAs are examined in terms of the precision along with the details of the implementation. The fast multipole moment (FMM) is today's fastest and best-scaling algorithm for large size problems. It is widely used in electrostatic and magnetic potential calculations, computations for stellar dynamics, even in molecular dynamics calculations, just to mention a few. The essence of the multipole method lies in the recognition that the relevance of single elements decreases with distance, consequently a group of farther-lying elements can be approximated as one (defined by multipole). The method requires to determine a radius for every cell, where direct computations shall be carried out only inside the radius. The further the elements are located from a certain cell the rougher approximation can be applied, i.e. larger and larger regions can be considered as a single multipole. In our current implementation we are modeling a micro magnetic array where the magnets are arranged on a  $N \times N$  grid. The magnets are located in fixed positions in the middle of the grid cells. In each step, the size of the grid is reduced to quarter by halving both width and height, and a new multipole is created in the middle of each  $2 \times 2$  sized cell. This process is repeated recursively until the resulting grid is reduced to  $4 \times 4$ . After the multipole representation is computed calculation of the field itself can be started from the coarsest level. Interactions between the current grid cell and grid cells outside of the cutoff radius on a given level are computed by using the multipole approximations, while interactions inside the cutoff radius are postponed to the finer level processing. The FMM methods, despite their advantage, have some implementation, calculation difficulty. They are highly memory intensive tasks and therefore the normal PC implementations are not so satisfying. FPGAs are highly parallel programmable logic devices with built-in memories thus an optimized calculation hardware can be realized with them. Our proposed architecture consists of customized processing blocks to compute the multipole representation and interactions inside and outside of the cutoff radius. The on-chip memory of the FPGA is used for the line buffers, which enables pipelined processing and the off-chip DDR main memory is used to store data for the different layers. In case of third order multipoles even a mid-sized FPGA can work on  $2048 \times 2048$  sized grid, which can be further increased by connecting multiple FPGAs together or by using larger FPGAs. Preliminary calculation shows that a mid-sized FPGA (XC7Z100) running at 200 MHz clock frequency can compute the above grid in less than 0.8 second. We believe that micromagnetic simulations, which are implemented on FPGA, open new perspectives in the simulation of magnetic and spintronic materials on widely different size scales.

### ***The role of charge transfer in the CO<sub>2</sub> Activation on 3d TM<sub>13</sub> clusters: A DFT investigation***

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Decreasing the concentration of CO<sub>2</sub> in the atmosphere is essential to mitigate the impact of global warming.<sup>1</sup> For that, one of the promising strategies is to convert CO<sub>2</sub> into higher value products through reduction; however, activating this molecule is difficult and also the conversion processes tend to be slow, being necessary the use of catalysts.<sup>1</sup> In this sense, from the computational point of view and using the Fritz-Haber ab initio molecular simulations package, this work provides insights of how CO<sub>2</sub> interacts and activates on 3d TM<sub>13</sub> clusters.<sup>2</sup> The magnitude of the adsorption energy,  $E_{ad}$ , indicates that the adsorption strength of CO<sub>2</sub> on the TM<sub>13</sub> clusters follows a periodic trend. In accordance to that, the C–O bond and OCO angle compared to CO<sub>2</sub> gas phase presented, respectively, an increase around 6.84 – 7.70% and a decrease around 22.96 – 24.32% for Fe<sub>13</sub>, Co<sub>13</sub>, and Ni<sub>13</sub>, while there were negligible structural modifications in the molecule for CO<sub>2</sub>/Cu<sub>13</sub>. Thus, as stronger as  $E_{ad}$ , higher is the geometric variations on the CO<sub>2</sub>, and vice-versa. Besides, the adsorption site preference in the lowest energy structure revealed that the CO<sub>2</sub> was coordinated in a bridge site on Fe<sub>13</sub>, Co<sub>13</sub>, and Ni<sub>13</sub> clusters, and in a top site on Cu<sub>13</sub> cluster. Hence, the molecule coordination is associated with its structural modifications, which permit to evaluate the activation of the molecule.

Also, to understand the character of the interaction between the molecule and the TM<sub>13</sub> clusters, we calculated the net charge at the molecule after the adsorption,  $\Delta q_{CO_2}$ . The results show that the CO<sub>2</sub> presents a partial negative charge when adsorbs on Fe<sub>13</sub>, Co<sub>13</sub>, and Ni<sub>13</sub>, indicating that those clusters donate charge to the molecule and, on the other hand, the  $\Delta q_{CO_2}$  on Cu<sub>13</sub> was found as almost zero. Furthermore,  $\Delta q_{CO_2}$  evaluates the grade of perturbation at CO<sub>2</sub> due to the adsorption, within those results being consistent with the  $E_{ad}$  and the CO<sub>2</sub> structural modifications, which allow us to classify the interaction as chemical adsorption on Fe<sub>13</sub>, Co<sub>13</sub>, and Ni<sub>13</sub>, and physical adsorption on Cu<sub>13</sub>, revealing the role of TM in the adsorption. Hence, the Fe<sub>13</sub>, Co<sub>13</sub>, and Ni<sub>13</sub> clusters promote the activation of CO<sub>2</sub> by the chemisorption, and the Cu<sub>13</sub> cluster favors its physical adsorption.<sup>2</sup>

[1] Esrafilzadeh, D.; Zavabeti, A.; et al. *Nat. Commun.*, 2019, 10(865), 1 – 8.

[2] Ocampo-Restrepo, V. K.; Zibordi-Besse, L.; Da Silva, J. L. F. *DFT Investigation of CO<sub>2</sub> Adsorption on 3d TM13 Clusters*. Manuscript in preparation.

### **Ring structure in two dimensional networks**

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The experimental realisation of ultra-thin materials such as amorphous graphene and silica bilayers has renewed interest in the structure two-dimensional (2D) atomic networks, first sparked by Zachariassen in 1932. These materials in fact form part of a wider class of 2D systems found throughout the natural world, which includes colloids, foams and even biological epithelia. Although these systems are physically very diverse, it has long been noted that they have strikingly similar structures, because they can all be viewed as a collection of percolating rings. Understanding the behaviours of 2D networks is therefore key many problems, from directed synthesis of nano-materials to controlling mitotic division to explaining curiosities such as the arrangement of the communes of Switzerland. We have developed multiple simple but physically motivated Monte Carlo methods to simulate 2D networks with a variety of morphologies and topologies. The first is based on the Wooten-Winer-Weaire algorithm for generating amorphous glasses, which has been modified to efficiently use the dual lattice; whilst the second is a modern computational variation of the approach used by Shackelford in his first hand-built models. We have used these simulations to precisely investigate the ring structure in network forming materials, helping to understand the origins of the empirical laws by which materials are currently characterised. These include Lemaitre's law, which describes the number of rings of a given size, and the Aboav-Weaire law, which describes the nearest-neighbour ring correlations. We will also discuss how the tools of modern network theory could be used to describe and investigate materials in the future.

### **Simultaneous sampling of multiple transition channels using adaptive paths of collective variables**

**Alberto Pérez de Alba Ortíz**, Bernd Ensing

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Enhanced sampling methods have made great progress expanding the observable time-scales in molecular dynamics (MD) simulations. Different types of schemes are nowadays widely exploited to study a vast variety of systems in chemistry, biophysics, material science, etc. In particular, biasing methods deliver insight in the form of free energy landscapes—with interpretable stable states, transition channels, intermediates and barriers—projected in the space of a few key molecular transition descriptors, i.e., collective variables (CVs). Complex systems, which require large sets of CVs to be described, pose an additional challenge: the convergence time of the free energy surface scales exponentially with the CV-space dimensionality. This challenge has been addressed by path-based approaches, which do not act on the CV-space itself, but instead on the 1D progress parameter along an adaptive curve connecting two known stable states in CV-space. In previous work, we have shown that path-metadynamics (PMD) is able to converge a transition path and the free energy along it with a sublinear rise in cost with respect to CV dimensionality. However, intricate systems often have multiple forking transition channels, which present additional hurdles for path-based methods, either by hindering the convergence of a particular path, or by conditioning the results to the initial guess. We introduce multi-PMD, a multiple-walker-multiple-path approach, which addresses this issue by means of introducing a controlled repulsion between the multiple paths. We showcase the performance of multi-PMD at the hand of clear case studies of short peptides, as well as a compelling application to the Watson-Crick-to-Hoogsteen base-pairing transition in DNA, which yields results agreeing to theory and experiment.

[1] G. Díaz Leines, and B. Ensing, *Phys. Rev. Lett.* **109**, 020601 (2012).

[2] A. Pérez de Alba Ortíz, A. Tiwari, R. C. Puthenkalathil, and B. Ensing. *J. Chem. Phys.* **149**, 072320 (2018).

[3] A. Pérez de Alba Ortíz, J. Vreede, and B. Ensing, in *Biomolecular Simulations: Methods and Protocols*, edited by M. Bonomi, and C. Camilloni (Springer, 2018) (In press).

[4] A. Pérez de Alba Ortíz, and B. Ensing, (In prep.).

## ***Aqueous solubility of sparingly soluble drug molecules and the role of hydrotropes*** **Sandip Paul<sup>1</sup>, Shubhadip Das<sup>2</sup>**

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Hydrotropes are an important class of molecules that enhance the solubility of an otherwise insoluble or sparingly soluble solute in water [1]. Besides this, hydrotropes are also known to self-assemble in aqueous solution and form aggregates [2]. It is the hydrotrope aggregate that helps in solubilizing a solute molecule in water [3]. In view of this, we try to understand the underlying mechanism of self-aggregation of different types of hydrotropes in water by using classical molecular dynamics (MD) simulation. We further extend our study to explore the hydrotropic action of these hydrotrope molecules towards the solubility of different sparingly soluble orally administered drugs in water [4-6].

[1] C. Neuberg, *Biochem. Z.* **76**, 107 (1916).

[2] S. Das, S. Paul, *J. Phys. Chem. B*, **119**, 3142 (2015).

[3] S. Das, S. Paul, *J. Phys. Chem. B*, **120**, 3540 (2016).

[4] S. Das, S. Paul *J. Chem. Inf. Model.* **57**, 1461(2017).

[5] S. Das, S. Paul *J. Phys. Chem. B* **121**, 8774 (2017),.

[6] S. Das, S. Paul *PLoS One* **13**, e0190209 (2018).

## ***A theoretical insight into the factors governing the specific binding within a ligand-receptor pair***

**Jasmina Petrova<sup>1</sup>, Gergana Gocheva<sup>1</sup>, Stoyan Iliev<sup>1</sup>, Nikoleta Ivanova<sup>2</sup>, Galia Madjarova<sup>1</sup>, Anela Ivanova<sup>1</sup>**

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Active targeting could be a highly efficient drug delivery approach that is nowadays attracting pronounced scientific attention. It may be put into action by employing the specific recognition of a drug carrier ligand by a membrane receptor overexpressed on the surface of malignant cells. In the present study, folate and a series of its structural analogues - antifolates, are considered as ligands of the folate receptor alpha, registered in the cells membranes of several types of cancer. The aim is to outline by molecular modelling the essential factors for the binding of the ligand, which stabilize its specific coupling to the receptor pocket. Quantum chemical calculations are applied for quantitative determination of the interactions taking place after the binding process and for deeper understanding of their nature. The computations are based on structures of the ligand-receptor complex obtained by spontaneous binding during atomistic molecular dynamic simulations. The results are discussed in terms of chemical variations of the ligands and of the supramolecular structure of the receptor.

## ***Range-separated density functional theory***

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In solid state physics, we encounter both IR and UV divergences in the evaluation of Feynman diagrams. These are related to the very nature of the Coulomb potential, namely its infinite range and the singularity at zero distance respectively. Even though the divergences are integrable, they drive up the cost of numerical methods significantly. Either sort of divergence can be cured by splitting the Coulomb potential in a long-range and complementary short-range part. Then, the part of diagrams that contains the divergence is approximated by density functional theory. I will discuss technical and theoretical aspects of range-separation techniques and talk about popular applications of the method such as the HSE functional. Furthermore, I will present findings of my own investigations into range-separated RPA.

## ***Large-scale biomembrane simulations based on a parametric model with realistic kinetics***

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We have developed a computationally efficient coarse-grained membrane model, to be used in concert with particle-based reaction-diffusion simulations [1]. The model is purely based on nearest-neighbor interactions between particles that each represent a coarse patch of a lipid monolayer. Interaction potentials are parameterized so as to reproduce the local membrane mechanics, whereas the in-plane fluidity is implemented with Monte Carlo bond-flipping moves. To tackle different kinetics of in-plane and out-of-plane degrees of freedom, and to achieve a realistic model of membranes suspended in solvents, we have developed an anisotropic stochastic dynamics scheme, based on exact solutions of Stokes equations. While drastically increasing the available sampling times, this approach also allows for modeling hydrodynamic interactions mediated by the solvent. Different aspects of the model are put to the test through studying thermodynamic as well as kinetic properties. Realistic kinetics of the model lead to correct prediction of dispersion relations in membrane patches, as well as large-scale dynamics of whole cells. We have also developed schemes to include interaction of the bilayer membrane with flexible curvature-inducing agents, resulting in quantitative predictions of membrane-mediated interactions and clustering of membrane-bound proteins. We expect this model to be of high practical usability in the context of ultra coarse-grained interacting particle reaction-dynamics (iPRD) simulations of biological systems [2].

[1] M. Sadeghi, T. R. Weigl, F. Noé, *J. Chem. Phys.* **148** 044901 (2018).

[2] M. Hoffmann, C. Fröhner, F. Noé, *bioRxiv* doi: 10.1101/374942 (2018).

## ***Towards exact molecular dynamics simulations with machine-learned CCSD(T) force fields and molecular properties***

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Molecular dynamics (MD) simulations employing classical force fields constitute the cornerstone of contemporary atomistic modeling in chemistry, biology, and materials science. However, the predictive power of these simulations is only as good as the underlying interatomic potential. Classical force fields based on mechanistic models of atomic interactions often fail to faithfully capture key quantum effects in molecules and materials. Here we enable the direct construction of flexible molecular force fields from high-level *ab initio* calculations by incorporating spatial and temporal physical symmetries into a gradient-domain machine learning (sGDML) model in an automatic data-driven way, thus greatly reducing the intrinsic complexity of the force field learning problem. The developed sGDML approach faithfully reproduces global force fields at quantum-chemical CCSD(T) and DFT{PBE0+MBD} level of accuracy and for the first time allows converged molecular dynamics simulations with fully quantized electrons and nuclei for flexible molecules and clusters with up to a few dozen atoms. Additionally, the combination of such results with molecular properties learned via continuous-filter convolutional neural networks (SchNet), allows the prediction of accurate spectroscopic properties. Our approach of learning highly accurate force fields and molecular properties provides the key missing ingredient for achieving spectroscopic accuracy in molecular simulations.



## **Simple wave-function in mean-field embedding for bulk systems using the plane-waves basis**

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Treating electronic correlation as a local phenomenon is an appropriate approach for many quantum chemical investigations. Local correlation effects also occur in bulk systems, e.g. in point defects or in molecule adsorption on a surface. It is therefore essential to limit accurate but expensive quantum chemical correlation methods (MP2, RPA, CCSD) to the local region of interest [1-3]. In this talk we demonstrate a simple scheme which partitions the electron density from a mean-field calculation into a local density and its complement. The occupied Bloch orbitals are unitarily rotated to localized orbitals by means of a singular value decomposition which maximizes the overlap between Bloch orbitals and atomic-like functions [4]. This approach preserves orthogonality between the orbitals forming the local density and its complement. While the mean-field energy captures all non-local electrostatic contributions, the effective system size for the correlation energy method is reduced to the local region. We demonstrate the potential of our embedding scheme by calculating the physisorption energy of a water molecule on a titanium dioxide surface.

[1] F. R. Manby, M. Stella, J. D. Goodpaster, T. F. Miller, *J. Chem. Theory Comput.* **8**, 2564 (2012).

[2] Gerald Knizia, Garnet Kin-Lic Chan, *J. Chem. Theory Comput.* **9**, 1428 (2013).

[3] I. W. Bulik, W. Chen, G. E. Scuseria, *J. Chem. Phys.* **141**, 054113 (2014).

[4] F. Libisch, M. Marsman, J. Burgdörfer, G. Kresse, *J. Chem. Phys.* **147**, 034110 (2017).

## **Towards multiscale modelling of the oxygen evolution reaction: DFT meets kMC**

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Photoelectrochemical (PEC) cells that produce hydrogen via the water splitting reaction are highly relevant in realization of a clean energy economy. The oxygen evolution reaction (OER) largely determines the energy efficiency of the PEC cells. To make OER energy efficient we need an in-depth mechanistic understanding of electrochemical processes at the complex solid-liquid interface which is a multiscale modelling problem [1]. While there are numerous computational mechanistic studies on OER over different materials, efforts towards multiscale modelling are in their nascent phase. In the present work, we have combined *ab initio* quantum chemical calculations (Density functional theory (DFT) and DFT based molecular dynamics (DFT-MD)) with **kinetic Monte Carlo** (kMC) simulations to elucidate the mechanism of the OER over hematite (Fe<sub>2</sub>O<sub>3</sub>) 110 surface at various electrode potentials. This is a first step towards developing a full multiscale model for the OER. We use the typical four step OER mechanism as proposed in [2]. Using kMC computations we simulate the surface coverage of the different species involved in the OER as a function of time and electrode potential together with the amount of evolved O<sub>2</sub>. Experimentally such detailed surface coverage data is difficult to obtain, however highly important to elucidate the OER mechanism. From the simulated kMC data, we can extract a qualitative current density vs electrode potential (**j-V**) curve which allows us to computationally determine the onset potential (Vonset) and make direct comparison with the experimental data. We found 1.44 V < Vonset < 1.46 V which is in good agreement with the experimental data [2b]. Future research will focus on detailed mechanistic analysis including solvent effects in DFT calculations and lateral interactions in kMC simulations.

[1] X. Zhang, A. Bieberle-Hütter, *ChemSusChem* **2016**, 9, 1223.

[2a] J. Rossmeisl, Z. W. Qu, H. Zhu, G. J. Kroes, J. K. Nørskov, *J. Electroanal. Chem.* **2007**, 607, 83.

[2b] F. L. Formal, M. Grätzel, K. Sivula, *Adv. Funct. Mater.* **2010**, 20, 1099.

## ***Computational grain boundary design of new magnetic materials: tuning the magnetization by impurity segregation***

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We present a systematic ab initio study of segregation of 12 non-magnetic sp-impurities (Al, Si, P, S, Ga, Ge, As, Se, In, Sn, Sb and Te) at  $\Sigma 5(210)$  grain boundary (GB) and (210) free surface (FS) in fcc ferromagnetic cobalt and nickel and analyze their effect on structure, magnetic and mechanical properties. We determine preferred segregation sites at the  $\Sigma 5(210)$  GB for the sp-impurities studied, their segregation enthalpies and strengthening/embrittling energies with their decomposition into the chemical and mechanical components. In nickel, most of the above impurities nearly kill or substantially reduce the magnetic moments at the FS and, when segregating interstitially (i.e. Si, P, S, Ge, As, Se), also at the GB so that they provide atomically thin magnetically dead layers which may be very desirable in spintronics. Reduction of magnetic moments at the  $\Sigma 5(210)$  GB in fcc ferromagnetic cobalt is, in absolute values, very similar to that in nickel. However, as the magnetic moment in bulk cobalt is higher, we do not observe magnetically dead layers here. It turns out that by focused impurity segregation we can generate atomically thin magnetic layers with tailored magnetization, which can contribute to a new development of technologically important materials.

Further, we use ab initio methods to assess tensorial elastic properties of interface configurations associated with GBs in intermetallics. Focusing on the  $\Sigma 5(210)$  GBs in Ni<sub>3</sub>Al compound as a case study, we evaluate the mechanical stability of the interface configurations by checking Born-Huang's stability criteria. The elastic constant C<sub>55</sub> is found three-/five-fold lower than in the bulk and, as a result, the mechanical stability of interface states is reduced. The tensorial elasto-chemical complexity of the interface configurations is demonstrated by a high sensitivity of elastic constants to the GB composition. In particular, we study elasticity changes induced by Si atoms segregating into the atomic layers close to the GBs and substituting Al atoms. If wisely exploited, our study paves the way towards solute-controlled design of GB-related interface configurations with tailored stability and/or tensorial properties.

Most of our results are theoretical predictions and we hope that they may motivate experimentalists to conduct new investigations in this field.

## ***Endohedral fullerenes for materials science***

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We use theoretical chemistry in predictive mode to understand chemical bonding and molecular properties of endohedral fullerenes hitherto unknown. We will present our study of X<sub>2</sub>@C<sub>n</sub> (X= F, Cl, Br; n=70, 80, 90) systems, in which the X<sub>2</sub> oxidizes fullerene cage while forming X<sub>2</sub>@C<sub>n</sub><sup>+</sup> systems with an electron occupying antibonding sigma<sub>u</sub> X-X molecular orbital and forming so-called single-molecule crystals. We will also discuss unwilling nature of actinide-actinide bonds formed in di-actinide fullerenes An<sub>2</sub>@C<sub>80</sub> (An=Ac--Cm), in which the bonds are formed to avoid charge repulsion between enclosed An ions. If time permits, we will show, how can be dipolar molecules inside C<sub>70</sub> manipulated by external electric field, and, how this, in combination with particular conductive properties of dipole@C<sub>70</sub> molecules, makes them candidates for molecular varistors and switching spin filters.

## ***A machine learning potential for hexagonal boron nitride***

**Fabian Thiemann**<sup>1,2</sup>, Patrick Rowe<sup>1</sup>, Erich A. Müller<sup>2</sup>, Angelos Michaelides<sup>1</sup>

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Hexagonal boron nitride (hBN), the inorganic equivalent of graphene, has gained in popularity due to remarkable properties such as high mechanical robustness, chemical stability and sorption performance [1,2]. Particularly, the application in desalination and water purification seems promising [3]. However, a

deeper understanding of the mechanisms behind the flow processes of fluids confined within BN-based nanostructures is required to understand the fascinating results shown by experiments [4,5].

While classical molecular dynamics (MD) is seemingly the ideal tool of choice, considerable doubts are cast about whether existing potentials accurately mimic the solid-fluid interactions [6-9] which strongly influence the behaviour at the nanoscale. To overcome this issue, new potentials are required to be built on a higher level of theory to account for local phenomena such as polarization. The emergence of machine learning (ML) has led to a paradigm shift in the development of force fields, where a massive amount of ab-initio data is processed providing a highly transferable and robust potential to faithfully represent the potential energy surface [10]. This offers the opportunity to expand the simulation time and systems size way beyond the density functional theory scale while retaining the accuracy of first principle methods for a fraction of the usual costs.

As a first step towards a sensible model for the solid-fluid interactions, an interatomic potential for hBN is constructed using the Gaussian approximation potential (GAP) [11] machine learning methodology. Later, we will extend our model towards systems comprising water which will then be used to perform adsorption and steady-state flow non-equilibrium MD.

- [1] A. Pakdel, Y. Bando, and D. Golberg, *Chemical Society Reviews* 43, 934–959 (2014).
- [2] W. Lei, D. Portehault, D. Liu, S. Qin, Y. Chen, *Nature Communications* 4, 1777 (2013).
- [3] G. Tocci, L. Joly, and A. Michaelides, *Nano Letters* 14, 6872-6877 (2014).
- [4] E. Secchi, S. Marbach, A. Niguès, D. Stein, S. Alessandro, and L. Bocquet, *Nature* 537, 210-213, (2016).
- [5] A. Pakdel, C. Zhi, Y. Bando et al., *ACS Nano* 5, 6507 (2011).
- [6] T. Hilder, R. Yang, V. Ganseh et. Al., *Micro & Nano Letters* 5, 150-156 (2010).
- [7] Y. Al-Hamdani, M. Ma, D. Alfè et al., *Journal of Chemical Physics* 142, 181101 (2015).
- [8] Y. Al-Hamdani, M. Rossi, D. Alfè et al., *Journal of Chemical Physics* 147, 044710 (2017).
- [9] J. G. Brandenburg, A. Zen, M. Fitzner et al., *Journal of Physical Chemistry Letters* 10, 358-368 (2019).
- [10] J. Behler, *Journal of Chemical Physics* 145, 170901 (2016).
- [11] A. P. Bartók, M. C. Payne, R. Kondor, and G. Csányi, *Physical Review Letters* 104, 136403 (2010).

### **Periodic coupled-cluster theory applied to molecule-surface interactions**

**Theodoros Tsatsoulis**, Andreas Grüneis

Institute for Theoretical Physics, Vienna University of Technology, Vienna - Austria

Over the last few years, quantum-chemical wavefunction based theories have been increasingly often applied to extended systems. In many instances these methods either exploit the locality of electronic correlation or are used within an embedding scheme. Here, we rely on a canonical periodic coupled-cluster theory implementation interfaced to the Vienna ab-initio simulation package (VASP). We show that the representation of virtual orbitals based on Gaussian basis-sets expanded in plane-waves[1], a low-rank factorization of the Coulomb integrals[2] and a finite-size correction scheme[3] allows for reducing the computational cost of the employed coupled-cluster methods. We demonstrate the capabilities of the methods by presenting results for water physisorption on two-dimensional C and BN sheets[4,5] and the energetics of hydrogen dissociation on the Si(100) surface[6].

- [1] G.H. Booth, T. Tsatsoulis, G.K. Chan, A. Grüneis, *J. Chem. Phys.* **145**, 084111 (2016).
- [2] F. Hummel, T. Tsatsoulis, A. Grüneis, *J. Chem. Phys.* **146**, 124105 (2017).
- [3] T. Gruber, K. Liao, T. Tsatsoulis, F. Hummel, A. Grüneis, *Phys. Rev. X*, **8**, 021043 (2018).
- [4] Y.S. Al-Hamdani, Y. S., *J. Chem. Phys.* **147**, 044710 (2017).
- [5] J.G. Brandenburg et al., *J. Phys. Chem. Lett.* **10**, 358 (2019).
- [6] T. Tsatsoulis, S. Sakong, A. Groß, A. Grüneis, *J. Chem. Phys.* **149**, 244105 (2018).

### **Molecular simulations of cellular membranes: coming of age?**

**Stefano Vanni**

University of Fribourg, Fribourg - Switzerland

I will present recent results from my lab showing how multi-scale molecular simulations have provided precise mechanistic insights into key cellular processes involving membranes, including vesicular trafficking (Vanni et al., *Nat Comm*, 2014; Magdeleine et al, *eLife* 2016), endocytosis (Pinot et al., *Science* 2014) and lipid droplet biogenesis (M'Barek et al., *Dev Cell* 201; Zoni et al, in preparation). I will highlight how recent developments have allowed to establish meaningful direct correlations between experimental observations and computational results, and I will discuss our efforts to develop next generation coarse-

grain force fields using machine learning and biophysical experiments to address important remaining key challenges in membrane simulations.

### ***Polaronic satellites in angle-resolved photoemission spectra from *ab initio* many-body calculations***

**Carla Verdi**

University of Vienna, Vienna - Austria

The coupling of electrons to bosonic excitations in solids, such as infrared-active phonons and carrier plasmons, has profound effects on the electronic properties of semiconductors and insulators. For example, bosonic coupling may result in the formation of polarons, i.e. electrons dressed by a boson cloud. Doped oxides constitute an exciting playground to investigate this phenomenon, as the polaronic nature of the charge carriers can strongly modify their optoelectronic properties.

In angle-resolved photoemission spectroscopy (ARPES) the signature of polarons is the appearance of spectral satellites below the conduction band. Experimentally, these features evolve into band-structure kinks with increasing doping concentration, challenging our understanding of the many-body interactions in these systems [1, 2]. Here we present our approach to calculate ARPES spectra from first principles by combining *ab initio* calculations of the electron-phonon and electron-plasmon coupling with the cumulant expansion method [3, 4]. This allows us to investigate polaronic quasiparticles and their evolution with doping from first principles. For the paradigmatic example anatase TiO<sub>2</sub>, we show that the transition from polarons to a weakly-coupled Fermi liquid with increasing doping observed in experiments originates from nonadiabatic polar electron-phonon coupling. A similar mechanism also applies to the ferromagnetic semiconductor EuO [5]. We observe that the coupling of electrons to low-energy carrier plasmons can sustain polaron formation analogously to the polar electron-phonon coupling. In particular, from combined ARPES experiments and *ab initio* many-body calculations we show that doped EuO can host plasmonic polarons with significant tunability with charge carrier doping.

[1] S. Moser *et al.*, *Phys. Rev. Lett.* **110**, 196403 (2013).

[2] Z. Wang *et al.*, *Nat. Mater.* **15**, 835 (2016).

[3] C. Verdi, F. Caruso, and F. Giustino, *Nat. Commun.* **8**, 15769 (2017).

[4] F. Caruso, C. Verdi, S. Poncé, and F. Giustino, *Phys. Rev. B* **97**, 165113 (2018).

[5] J. M. Riley *et al.*, *Nat. Commun.* **9**, 2305 (2018).

### ***Investigation of kinetic functionals applied to metal nanoparticles***

**Urso Vittoria**<sup>1</sup>, Fabio Della Sala<sup>1,2</sup>, Eduardo Fabiano<sup>1,2</sup>, Lucian Constantin<sup>1</sup>

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I develop advanced kinetic energy (KE) functionals for orbital-free (OF) density functional theory (DFT) applications. The KE functionals developed have tested on a newly developed benchmark which extend previous benchmark sets with bulk solids, kinetic potentials for OF-DFT, and absorption spectra of jellium systems via the hydrodynamic approach. I present a study of Laplacian-Level Kinetic Energy functionals applied to metallic nanosystems. The nanoparticles are modeled using jellium spheres of different sizes, background densities, and number of electrons. The ability of different functionals to reproduce the correct KE density and potential of various nanoparticles is investigated and analyzed in terms of semilocal descriptors. Most semilocal KE functionals are based on modifications of the second-order gradient expansion (GE2) or fourth order gradient expansion (GE4). Gradient expansions are in fact powerful theoretical tools which describe with accuracy the KE slowly varying density regime of an electronic system, providing an ideal starting point for the development of approximate KE models.

### ***Expedite random structure searching using objects from a Wyckoff position***

**Ching Ming Wei**, Cheng-Rong Hsing

Institute of Atomic and Molecular Sciences, Academia Sinica, Taipei - Taiwan

Random structure searching [1] has been proved to be a powerful approach to search the global minimum and the metastable structures. A true random sampling is in principle needed yet it would be highly time-consuming and/or practically impossible to find the global minimum for the complicated systems in the

high-dimensional structural space. Thus the implementations of reasonable constraints, such as adopting system symmetries to reduce the independent dimension in structural space and imposing chemical information to reach and relax into low-energy regions, are the most essential issues in the approach. In this talk, we propose the concept of "object" [2] which is composed an atom or a set of atoms (such as molecules or carbonates) carrying symmetry defined by one of the Wyckoff letters and thus led to the searching of global minimum for a large system confined in a greatly-reduced structural space is accessible in practice. We will examined several representative materials including solid methanol, high-pressure iron carbonates ( $\text{FeCO}_3$ ), fullerene ( $\text{C}_{60}$ ),  $\text{Si}(111)\text{-}7\times 7$  dimer-adatom stacking faulted reconstructed surface, and high-pressure manganese carbonates ( $\text{MnCO}_3$ ) [3] to demonstrate the power and the advantages of using "object" concept in random structure searching.

1. C.J. Pickard, and R. J. Needs, *J. Phys.: Condens. Matter* **23**, 053201 (2011).

2. S.W. Wang, C.R. Hsing, and C.M. Wei, *J. Chem. Phys.* **148**, 054101 (2018).

3. H.C. Hung, C.R. Hsing, and C.M. Wei, *Phys. Rev. B* **99**, 024106 (2019).

### ***Partial dissociation of antigenic peptides from mhc i or how to deal with conflicting results from different enhanced sampling methods?***

**Sebastian Wingbermühle**, Lars Schäfer

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Major Histocompatibility Complex I (MHC I) is one of the key players in adaptive immunity. Expressed on the surface of all nucleated cells, it displays sample antigenic peptides from the cytosol to patrolling cytotoxic T-cells that can thus identify and kill malignantly transformed and virally infected cells. Although several crystal structures of MHC I in complex with an antigenic peptide (pMHC I) have been solved, the structural dynamics of pMHC I at the cell surface remain largely elusive. According to a recent NMR study on HLA-B\*35:01[1], pMHC I complexes can adopt a minor state in which the antigenic peptide is bound tightly to MHC I and a major state in which the peptide is bound more loosely. In unbiased MD simulations of the pMHC I studied by Yanaka et al., the peptide N-terminus dissociated from the MHC I binding groove during a few hundred nanoseconds. This finding suggests that pMHC I with completely bound peptide may correspond to the proposed minor state, whereas pMHC I with partially dissociated peptide may constitute the major state. To underpin this, the potential of mean force (PMF) along the distance between the anchor residue of the peptide N-terminus and its binding partner in the MHC I binding groove has been calculated. Results from bias exchange simulations (1 microsecond of MD simulation per umbrella window) suggest that pMHC I in which the peptide N-terminus has dissociated from MHC I are lowest in free energy. Contrarily, replica exchange with solute tempering (REST2) simulations (2 x 2 microseconds), in which the effective temperature of the MHC I binding groove and the antigenic peptide is increased, predict pMHC I with completely bound peptide to be the global minimum of the free energy. This seemingly contradicting difference is discussed and further elucidated.

[1] Yanaka et al., *J. Biol. Chem.* (2014).

### ***Influence of polar co-solutes and salt on the hydration of lipid membranes***

**Amanuel Wolde Kidan**<sup>1</sup>, Quoc Dat Pham<sup>2</sup>, Alexander Schlaich<sup>3</sup>, Emma Sparr<sup>2</sup>, Roland Netz<sup>1</sup>, Emanuel Schneck<sup>4</sup>

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<sup>4</sup>Max Planck Institute of Colloids and Interfaces, Potsdam - Germany

Lipid membranes form the diffusional barrier of eukaryotic cells and determine all processes where cells come into close contact, for example during cell fusion or vesicle formation. In this work, the influence of the co-solutes TMAO, urea, and NaCl on the hydration repulsion between lipid membranes is investigated in a combined experimental/simulation approach. Pressure-hydration curves reveal that the repulsion significantly increases when the membranes are loaded with co-solutes, most strongly for TMAO. As a result, the co-solutes retain additional water molecules and therefore provide membranes with a fluid and more physiological environment. This describes a hitherto neglected aspect of the "fluidizing" action of naturally occurring osmolytes. Experimental calorimetry data are quantitatively reproduced in

complementary solvent-explicit atomistic molecular dynamics simulations, which yield the chemical potential of water. Simulation analysis reveals that the additional repulsion arises from the osmotic pressure generated by the co-solutes, an effect which is maximal for TMAO, due to its unfavorable interactions with the lipid headgroup layer and its extraordinarily high osmotic coefficient.

### ***Elasticity of randomly diluted fibre networks***

**Sina Zendehroud**, Roland Netz

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The basement membrane, a network of proteins spanning over the interface of the epithelium and tissue, has been of increased interest lately because of its crucial role in metastasis of cancer cells. Acting as a mechanical barrier, it prevents malignant cells from invading deeper tissue. Thus, the malignancy of carcinoma is often directly related to their ability to break through the basement membrane. Inspired by recent experiments on basement membrane-like matrices, we study theoretically the effects of site percolation on the elasticity of fibre networks. Using an in-house code, simulations of the elastic network at different degrees of dilution are performed, determining the elastic response by applying a longitudinal deformation. We find that even at very low degrees of dilution, the material behaves like a two-phase system, not responding at all to strains below a critical strain, and responding linearly above that value. Using this result, we are able to construct an effective model successfully predicting experimental measurements on bulk samples of diluted basement membrane-like matrices.

# A European Research Centre in Orsay: Cecama 1969-1993

Giovanni Battimelli  
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## A EUROPEAN RESEARCH CENTRE IN ORSAY: CECAM 1969-1993

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### Gestation and birth

Half a century after its creation, CECAM (Centre Européen de Calcul Atomique et Moléculaire) is today a solid research institution with a complex and articulated structure. Its official mission is “the promotion of fundamental research on advanced computational methods and their application to important problems in frontier areas of science and technology”. With headquarters in Lausanne, where the Director, Deputy Director and the administrative staff are located, it spreads across Europe and Israel through a network of 17 operating centres (“nodes”), and is sponsored by 25 research organisations from 14 different European countries. All member organisations are represented in the Council, CECAM’s supreme governing body, while the implementation of the scientific activities is controlled by a Board, chaired by CECAM’s Director, whose members are the Node Directors. A Scientific Committee acts as advisor to the Board on the CECAM scientific policy.

Nothing of the sort was in sight when CECAM came into existence. Contrary to other international research institutions (of whom CERN, the European laboratory for high energy physics created in Geneva in 1954, probably represents the best and more widely known example), that were established by official agreements between the research institutions, if not the governments, of the countries involved, CECAM was at the beginning (and stayed for quite some time after) a kind of informal organisation which managed to operate successfully in spite of lacking a clearly defined institutional status. Often behind any such large cooperative enterprise are the vision and determination of a few individuals and this is particularly true in the case of CECAM, which has often been depicted as a “family affair”. As a family, CECAM grew rapidly to become a large one; but it was always clear to every member of the family who the *pater familias* was. And, irony of history, the father who gave birth to the most successful European enterprise in the field of computational science was an American.

Born in Terre Haute, Indiana, in 1922, Carl Moser had received his PhD in organic chemistry at Harvard University in 1948, followed by a position as assistant professor at Johns Hopkins. In 1951 he moved to England, with a postdoctoral fellowship in theoretical chemistry, to improve his knowledge of quantum mechanics with Charles Coulson in London and Oxford. On Christmas, in 1952, he visited Paris, and got acquainted with Raymond Daudel,



the director of the Centre de Chimie Théorique de France, that Daudel had founded in 1944. It was the first and main research centre in quantum chemistry in the country, which later (1954) changed name in IMOACR (Institut de Mécanique Ondulatoire Appliquée à la Chimie et à la Radioactivité), and finally became in 1957 CMOA (Centre de Mécanique Ondulatoire Appliquée), a laboratory of the CNRS.<sup>1</sup> Daudel invited Moser to look forward to a career in CNRS, and to move from England to his centre for quantum chemistry. Moser accepted, and settled in Daudel's centre in the fall of 1953, thus becoming "an American in Paris". By the mid-sixties Moser had climbed the steps of the CNRS ladder, attaining the post of Directeur de Recherche and leading one of the four large research groups of CMOA, which in 1962 had moved from its previous location in rue de Sèvres to a new housing at 23 rue du Maroc, to take advantage of the facilities of the computing centre of the Institut Blaise Pascal, operating at the same place.<sup>2</sup>

In his first ten years or so in Paris, Moser established a dense network of relations throughout Europe with scientists and scientific managers in quantum chemistry and related fields, became quite dissatisfied with his own research, that "would never lead to a Nobel Prize"<sup>3</sup>, and had reached the conclusion that the future advancements in his science would require large investments in computing facilities. It was, in hindsight, an easy prediction to make in the mid-sixties, given, on one side, the ongoing development of computers and, on the other, the growing need for numerical efficient techniques required in those fields, such as atomic and molecular chemistry and physics, where advances on purely analytical grounds were prevented by computing difficulties. It was still easier, for someone familiar with the state of the art in computer science and its application to fundamental research, to evaluate the extent of the gap existing at the time between the flourishing developments in the U.S. A. and the still very limited and poorly exploited availability of similar resources throughout Europe. The project of setting up some kind of cooperative effort to strengthen European activity in computational science thus started to take shape.

The idea must have been floating around for a while. From the scarce and fragmentary evidence available, it seems that it was openly debated for the first time at a meeting on molecular physics held in Blaricum (Holland) at

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<sup>1</sup> About CMOA, and more general developments in France, see J.L. Rivail, B. Maigret, *Computational Chemistry in France: A Historical Survey*, Reviews in Computational Chemistry 12, 1998, pp. 367-380.

<sup>2</sup> Created in 1946 as a centre for applied mathematics, the Institut Blaise Pascal was one of the main institutional actor in the development of computer science in France throughout the 60's. See A. Collinot, P.E. Mounier-Kuhn, *Forteresse ou Carrefour: l'Institut Blaise-Pascal et la naissance de l'informatique universitaire parisienne*, La Revue pour l'Histoire du CNRS, 27-28, Automne-Hiver 2010, pp. 79-88.

<sup>3</sup> M. Karplus, *Carl Mathew Moser*, Carl Moser Symposium, CECAM, Lyon 2005.

the end of March 1967, “under the leadership of C.M. Moser”.<sup>4</sup> Blaricum was the seat of the European Education Center, founded in 1959 by IBM World Trade Corporation, a subsidiary of IBM focused on foreign operations. Nothing else is known about this meeting that is however always referred to as the moment in which the very idea of what was to be CECAM materialized. The most detailed account comes from Wim Niewpoort, professor of Theoretical Chemistry at the University of Groningen: “in 1967 Carl invited me to join his famous “Blaricum meeting”, at an IBM center. There he exposed his idea about a European center for atomic and molecular calculations to a critical but knowledgeable audience including Bob Nesbet, Enrico Clementi, Paul Bagus, Gerd Diercksen, Brian Sutcliffe to mention a few. The reception was mixed. I remember my own reaction. As a true Dutchman I favoured a careful start through bilateral cooperation, rather than an all-out multilateral organization.”<sup>5</sup> A bit more information is given by Jan Mulder, then still working at his doctoral dissertation at Leiden University: “Carl Moser came into my life at the Blaricum meeting, 29-31 March 1967. Rather early in the course of my thesis research I had started applying computational methods... and so the possible creation of a European Institute for computational physics and chemistry seemed a highly interesting development. I remember meeting Carl again in London at the Faraday Symposium “Molecular Wavefunctions”, 12-13 December 1968... It had become clear that CECAM would start in the fall of 1969 and between my thesis supervisor L.J. Oosterhoff and C.M. Moser it was decided that I should go to Orsay (as it would turn out) to learn ab initio quantum-chemical calculations.”<sup>6</sup>

Indeed, by December 1968 it was clear that something would materialize. Soon after the Blaricum meeting Moser had presented a project to the high officers of CNRS. His plan was to link his initiative with the decision taken by CNRS Director General Pierre Jacquinot to create a new large computing centre (incorporating the old centre of the Institut Blaise Pascal, which was going to be dissolved). Moser’s project was first presented by Hubert Curien (which would replace Jacquinot as Director General in 1969) at a meeting of the Comité de direction of the CNRS, on September 14, 1967: the project aimed at “regrouping the researchers working on quantum computations in order to realize programs competitive with those in the United States”; it was stated that a meeting recently held in Holland had given indication that such a cooperation was desirable and feasible, that “Germany, Great Britain, Italy and maybe the USSR might give a contribution of 4 – to 500.000 Francs each”, while France could participate by offering computer time

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<sup>4</sup> The only reference I could locate in the scientific literature to the Blaricum Meeting, apart from personal recollections by some participants, is in *Proceedings of the Thirteenth General Assembly, Prague 1967*, International Astronomical Union, 1968, p. 87.

<sup>5</sup> W. Niewpoort, in *Recollections of CECAM – For Carl*, CECAM, Orsay 1990, p. 23.

<sup>6</sup> J.C. Mulder, in *Recollections of CECAM – For Carl*, CECAM, Orsay 1990, p. 21.

and paying researcher's salaries; the new Institut would be centred around the computer that CNRS intended to acquire.<sup>7</sup>

No formal decision was taken at the time. A consensus was reached one year later. In the meantime, Jacquinot's project of a new large computing facility had materialized, and work was going on at the site of Orsay to install there the new-born Centre Inter-Régional de Calcul Électronique (CIRCÉ), which was officially inaugurated on January 1969. At the meeting of the Comité de Direction held on October 7 1968, it was agreed that CNRS would support Moser's project; for the first time the acronym CECAM appeared in an official document. The minutes of the meeting relate that "since three years" contacts had been established by Moser to create a European centre on atomic computation; that the best specialists from Europe and America were expected to take part in its activities; that British, Dutch, Italians, Germans and French scientists were already showing a deep interest in the proposal (the USSR had in the meantime disappeared); that important firms such as IBM were willing to give their contribution. Moser made three requests to CNRS: computing time on the 360-75 (the IBM machine to be installed at CIRCÉ); lodging for his Centre next to CIRCÉ on the site of Orsay; an (unspecified) amount of money to cover the expenses of visiting scientists. The majority of the Committee's members "were seduced by the perspective of having an international laboratory of high scientific quality", but objected to the idea that CNRS should pay for the visits of foreign visitors. These expenses, it was agreed, should be covered by their respective countries.<sup>8</sup>

Therefore, by the end of 1968 the CNRS agreed to start the project, but it was not clear what formal status the new Centre would be given.<sup>9</sup>

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<sup>7</sup> "M. CURIEN expose le projet de M. MOSER tendant à regrouper les chercheurs spécialisés dans les calculs quantiques, afin de mettre au point des programmes susceptibles de rivaliser avec ceux qui sont montés aux Etats-Unis. Une première réunion de ces chercheurs, qui a eu lieu récemment en Hollande, a montré que les possibilités de cette coopération sont réelles. M. MOSER propose de regrouper cet institut autour de l'ordinateur que le C.N.R.S. a l'intention d'acheter. Il estime que le budget de fonctionnement s'élèverait aux environs de 6 (?) millions de francs pour une vingtaine de personnes. L'Allemagne, la Grande-Bretagne, l'Italie et peut-être l'U.R.S.S. pourraient y contribuer à raison de 4 à 500.000 F pour chaque pays. La France pour sa part, y participerait par la fourniture d'heures-machine et le paiement des chercheurs." Extrait du compte-rendu du Comité de direction n° 56 - Séance du jeudi 14 septembre 1967 - Point n° 6 - Projet d'institut européen de recherches moléculaires, CNRS Archives, Dépôt de Gif-sur-Yvette, versement 910001.

<sup>8</sup> Extrait du compte-rendu du Comité de direction n° 157 - Séance du lundi 7 octobre 1968 - Point n° 4 - Centre de recherche européen de calcul atomique et moléculaire (C.E.C.A.M.), CNRS Archives, Dépôt de Gif-sur-Yvette, versement 910001.

<sup>9</sup> Several holes in the documentation preserved both in Lausanne and at the CNRS archives regarding the early stages of CECAM make it difficult a detailed reconstruction of events, and offer sometimes puzzling evidence. Such is the case of a "CECAM brochure", found among other scattered documents in the CNRS archives. Only pages 7, 11 and 13 have been photocopied and preserved. Since the first pages are missing, the exact date of the document

Disregarding such bureaucratic subtleties, Moser went ahead, and by October 1969 CECAM was installed at its new premises on the hill in Orsay; that is, Moser moved himself, his office, his secretary and his beloved dogs from rue du Maroc to a room in the Laboratoire Aimé Cotton (the CNRS laboratory for atomic spectroscopy), located at Batiment 505, next to the building where CIRCÉ was installed. Not much later (when exactly we cannot say) CECAM crossed the street and settled finally in Batiment 506, where enough space for its activities was afforded by the CIRCÉ director, Janine Connes. Connes, an astrophysicist who had acquired a solid competence in computational science working in 1963 at the Jet Propulsion Laboratory in Pasadena, had been entrusted by Jacquinot to be responsible of the management of the new large computing centre at Orsay; although completely foreign to the field of quantum chemistry, she shared with Moser the feeling that a strong effort was needed from Europe to become competitive with the U.S. in computational science, and was sympathetic with his plans for such a cooperation. She therefore proposed to Jacquinot to host Moser's new centre in the CIRCÉ's building where (at least for the moment) space was available and not yet fully utilized.<sup>10</sup>

## A European network

“How he did it I do not know, but in 1968 [must be 1969] I got word that an organization called CECAM was brought into existence under his

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is unknown (although from its content it could be located around the beginning of 1969), nor is it possible to ascertain its author (which may well be Moser himself). It is announced that CECAM will be opened at the CNRS Computing Centre in Orsay in October 1969. About twenty scientists are expected to visit the Centre for a long period of time (hopefully “a year at least”), mostly but not exclusively from Europe. Mention is made of a “Comité de Direction Européen” that should determine the amount of the financial contribution from each foreign institution taking part in the Centre's activities. Most interesting is a list of scientists (all of them active in quantum chemistry) to whom aspiring visitors to CECAM are addressed to establish contacts: besides Moser, we find for France also R. Chabbal from the Aimé Cotton CNRS laboratory at Orsay, W.A. Bingel and W. Kutzelnigg (both from Göttingen) for Germany, for Britain D.W. Cruikshank from Manchester and J.W. Linett from Cambridge, and Italians M. Simonetta from Milan and A. Vaciano from Rome. The closing lines of the booklet stress the provisional form of the Centre, that will be “certainly closed five years from now, should it prove to be a failure”; while “if, as we strongly believe, it will be a success, it will be necessary to set up an European Organisation for its administration”. CNRS Archives, Dépôt de Gif-sur-Yvette, versement 910001.

<sup>10</sup> The information that hosting CECAM in Batiment 506 was a suggestion by Janine Connes was given to me by Mme Connes herself, in a short conversation we had on June 13, 2019 in the office of IDRIS Director Denis Giroud, in that very Building 506 at Orsay (IDRIS is the new computing centre that took the place of CIRCÉ in 1994). That the first space occupied by CECAM in Orsay was at the Laboratoire Aimé Cotton is confirmed by Carl Moser in his introduction to the 1978 CECAM Scientific Report (written in 1979): “I would not wish to finish this introduction without a reminder that on October 1 of this year ten years will have passed since we started operation in the Laboratory Aimé Cotton here in Orsay”.

directorship and located at Batiment 506, Campus d'Orsay. Interested scientists were invited for research stays during which they could use computational facilities of CIRCÉ with generous limits."<sup>11</sup> No doubt, "how he did it" was largely due to Moser's determination, and disregard of bureaucratic matters. In dealing with scientific administrators, Moser displayed a pragmatic attitude coupled with a distinctive tendency to "think big" (and ask consequently). "Above all, Carl had the streak of outrageousness, which was essential in setting up a major facility and convincing governmental bodies. His first tool in these enterprises was making shameless demands. At CMOA he asked for an annual budget of computer times of 1000 hours, while the rest of the department asked for 20! He got 800."<sup>12</sup>

While CECAM started operating on scientific grounds, its existence at the formal level was still a matter of embarrassment for high CNRS officials. It was difficult indeed to find an adequate way to give official life to an institution that consisted only of a Director and a secretary, with no permanent personnel. Already at the 1967 meeting where Moser's project was first discussed it had been decided to avoid the creation of "an independent moral figure" to define the legal status of the proposed centre, and it was rather suggested to have it in some way directly supported on formal aspects by CNRS.<sup>13</sup> Exactly which way, however, was left undefined, and such was the situation when CECAM moved to Orsay. A first suggestion to establish the centre under the form of a Recherche Coopérative sur Programme (RCP) was abandoned, just as the idea of creating it as an association following the lines of the 1901 Loi des

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<sup>11</sup> W. Nieuwoort, in *Recollections of CECAM – For Carl*, CECAM, Orsay 1990, p. 23.

<sup>12</sup> G. Richards, in *Recollections of CECAM – For Carl*, CECAM, Orsay 1989, p. 25. Richards also mentions a second strategic move used by Moser to have his requests promptly accepted, which though anecdotic deserves to be reported: "The second line of attack on ministers and hautes fonctionnaires was Gunther. Gunther was Carl's child – a dachshund who had a spinal injury and was in consequence paraplegic. Carl very much judged people by their reactions to Gunther, whom he loved and cared for indefatigably. Being a spastic, however, Gunther had no control over his bladder. Thus when Carl went to see the minister to demand resources for CECAM he took Gunther into the carpeted offices, and many of us felt that funds were often forthcoming because officials were worried about their carpets and gave in to Carl to get him out of their suites."

<sup>13</sup> "Quant à la forme juridique de cet institut, M. DAURY (?) souhaiterait éviter la création d'une personne morale indépendante. Aussi est-il décidé par le comité de proposer à M. MOSER une formule qui ferait du C.N.R.S. le support juridique de cet organisme sous la forme d'une R.C.P. européenne assortie d'apports étrangers régis comme les ressources affectées. Le comité propose aussi de modifier la dénomination de cet institut, le terme de « recherches moléculaires » lui paraissant un peu large pour un objet qui est en réalité assez spécialisé." Extrait du compte-rendu du Comité de direction n° 56 - Séance du jeudi 14 septembre 1967 - Point n° 6 - Projet d'institut européen de recherches moléculaires, CNRS Archives, Dépôt de Gif-sur-Yvette, versement 910001.

Associations.<sup>14</sup> On December 12 1969, CNRS Director General Hubert Curien wrote to the Director of cultural, scientific and technical relations at the Department of Foreign Affairs, on the general subject of European cooperation, raising the issue of how to deal with the legal status of CECAM. Curien underlined the specific difficulty of the problem: it was neither the case of a cooperation between two (or more) laboratories from different countries, that could have been easily defined through bilateral conventions or the application of the existing formula of the RCPs, nor of a large European or international project requiring for its implementation the official ratification of complex intergovernmental agreements. The case at hand (“the creation of a European centre for atomic and molecular computations, that could be installed next to the CNRS computing centre at Orsay”) was of an intermediate size, making a formal intergovernmental agreement unnecessary, but still requiring “a formal structure to guarantee coordination of research and proper operation handling”. For the time being, Curien concluded, CNRS would find a provisional way of handling the matter, but, should the initiative be successful, a “more definitive solution should be found in the forthcoming years”.<sup>15</sup>

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<sup>14</sup> The CNRS files keep a copy of the (undated, but clearly end of 1969) draft of a document “Statuts du Centre Européen de Calculs Atomiques et Moléculaires”. CECAM was there conceived as a private association among individuals, having as founding members R. Chabbal, H. Curien and C. Moser for France, and V. Caglioti, M. Simonetta and A. Vaciago for Italy. The document had been prepared for approval from the competent CNRS offices, and rejected on administrative grounds, as stated by H. Curien in his letter on the matter cited in note 14. Most likely the rebuttal of the proposal had to do with the fact that in the Statute of the proposed association no mention at all was made of any role of CNRS, and the question of how the financial aspects would be handled was left totally undefined.

<sup>15</sup> In view of its interest, we reproduce the full text of the letter: “Décembre 12, 1969 - Le Directeur General du CNRS à Monsieur le Directeur General des relations culturelles, scientifiques et techniques, Ministère des Affaires étrangères - Objet: Coopération européenne.

Au cours d’une récente conversation, nous avons évoqué le développement nécessaire d’actions européennes en matière de recherche scientifique et l’insuffisance actuelle des moyens juridiques permettant de promouvoir de telles actions.

Lorsqu’il s’agit d’une collaboration directe entre deux laboratoires, l’un français, l’autre étranger, chacun apportant ses moyens à l’exécution du programme, aucune difficulté ne se présente. Cette collaboration peut s’inscrire soit dans le cadre des protocoles d’application des conventions de coopération scientifique et technique relevant de votre Département, soit dans celui des conventions passées entre le C.N.R.S. et certains organismes nationaux de recherche étrangers.

Dans le même esprit, quelques laboratoires étrangers participent avec des laboratoires français à l’exécution de “recherches coopératives sur programme”. Il s’agit, ici encore, de rapports directs entre laboratoires sur des sujets de recherche fondamentale. Cela ne soulève pas de problème particulier et cette formule pourrait sans doute être développée à l’avenir.

Par ailleurs, s’il s’agit de projets européens ou internationaux de grande envergure, il est normal que ces projets fassent l’objet d’accords entre gouvernements, mais la procédure est généralement assez lourde et demande des délais assez longs.

From the available documentation it seems that CNRS took a decision about the “provisional way of handling the matter” on January 14 1970, creating “a specific action for the opening of the Centre européen de Calcul atomique et moléculaire”.<sup>16</sup> The final arrangement left to the CECAM director full autonomy for the scientific management of the centre, while the financial issues connected with the sponsoring of CECAM activities by foreign partners would be regulated by specific conventions between those partners and CNRS, which would operate as administrator of CECAM related funds. In particular, Moser insisted (and this line of action was strictly enforced, despite episodic objections from some of the member institutions) that funds allocated by the different partners should go into the general budget for CECAM activities, and not exclusively used to cover the expenses of the scientists from that specific country. By so doing, Moser intended to leave open the possibility to use CECAM money for all sort of general scientific activity and eventually allow participation to those activities also for researchers coming from countries whose institutions were not official CECAM partners. Benefit for the financing countries would come mostly from having their scientists taking advantage of the computing facilities of CIRCÉ and interacting with a broadly international network of colleagues.

Contacts with various national agencies were soon established through the thick network of Moser’s personal relations. Wim Niewpoort acted as the “CECAM connection man” with Holland: “Carl and I did our best to gain support for a formal agreement with CECAM in Holland. In 1970 I went to Orsay for three months with the specific aim to prepare a fact-finding report and advice ZWO, the Dutch science foundation. Shortly after negotiations began

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Le problème qui nous préoccupe est celui d’actions intermédiaires de moindre envergure pour lesquelles il ne paraît pas indispensable de conclure un accord inter-gouvernemental, mais qui nécessitent une assise juridique pour assurer la coordination des recherches et la gestion des opérations.

En voici un exemple : celui de la création d’un Centre européen pour des calculs atomiques et moléculaires, pouvant être implanté près du Centre de calcul du C.N.R.S. à Orsay.

Nous avons pensé initier cette entreprise en constituant une association de la loi de 1901, mais cette solution n’a pas reçu l’agrément de notre Contrôleur financier. Nous pouvons amorcer la coopération grâce à une action spécifique du C.N.R.S., mais il nous faut trouver, si l’expérience se révèle concluante, une solution plus définitive dans les années à venir.

Je vous serais reconnaissant si, ainsi que vous me l’avez proposé, il vous était possible de faire étudier ce problème par vos services.”

It is interesting to note that, writing on December 12, Curien says that CECAM “could be installed” at Orsay, while in fact it had actually already been operating there for almost three months.

<sup>16</sup> Explicit reference to the “decision to create a specific action” taken on January 14, 1970, is made in the Convention between CNR and CNRS of May 12, 1970. No document allowing to know what exactly was decided on that day could be found either in the CNRS or in the CECAM archival files. It is also impossible to establish when, and by which body, Moser was officially designated as the CECAM Director.

that led to the participation of ZWO (now NWO) in CECAM, which still exists today.”<sup>17</sup> André Bellemans, professor of quantum chemistry at the Libre Université de Bruxelles, and Georges Verhaegen, who had worked for his PhD thesis at CMOA (making good use of the vast amount of computer time that Moser had asked and obtained), played the same role with the Belgian FNRS (Fonds National de la Recherche Scientifique).<sup>18</sup> The “Convention de recherche” between CNRS and the Italian Consiglio Nazionale delle Ricerche (CNR) was signed on May 12 1970, by CNR President Vincenzo Caglioti and the CNRS Administrative Director Pierre Creyssel; it was intended to last for three years, and was indeed renovated on January 1 1973.

CECAM activities in 1971 were directly supported by CNRS (France), CNR (Italy), ZWO (Holland), FNRS (Belgium); in addition, further grants were received from the French Centre d’Etudes de Limeil of the CEA (Commissariat à l’Energie Atomique) and from IBM France and Control Data (France).<sup>19</sup> Control Data soon disappeared, and in the following years the group of the sponsoring partners remained essentially the same: the four national scientific agencies of France, Italy, Holland and Belgium (CNRS, CNR, ZWO, FNRS), CEA-Limeil, and IBM France. They were joined in 1975 by the Belgian Ministère de l’éducation nationale et de la culture française (followed in 1981 by its Flemish counterpart, Ministerie van Nationale Opvoeding en Nederlandse Cultuur), the University of Namur (Belgium), the Centre d’études nucléaires de Saclay (CEA, France) and, for a period of two years only, the Délégation générale à la recherche scientifique et technique (DGRST, France). The Namur University withdrew in 1980, and so did IBM France the following year. 1981 saw the arrival of direct funding from the main British agency, the Science Research Council (SRC), that changed name in 1982 to become SERC (Science and Engineering Research Council). Despite this late official

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<sup>17</sup> W. Niewpoort, in *Recollections of CECAM – For Carl*, CECAM, Orsay 1990, p. 23. The Nederlandse Organisatie voor Zuiver-Wetenschappelijk Onderzoek (ZWO) was the Dutch equivalent of CNRS, established in 1950, whose activity was limited to pure science; in 1988 it was renamed Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO), covering both pure and applied science.

<sup>18</sup> The Belgian National Fund for Scientific Research (NFSR) is split in two separate organisations: the Fonds Wetenschappelijk Onderzoek – Vlaanderen (FWO), for the Flemish Community, and the Fonds de la Recherche Scientifique – FNRS (F.R.S. – FNRS) for the French-speaking part of Belgium. In the CECAM Scientific Reports, they are usually designated as FNRS.

<sup>19</sup> This, and most of the information that follows, is taken from the yearly CECAM Scientific Reports. The CECAM archives in Lausanne preserve an almost complete series of these Reports. Unfortunately, no copy is left of the first Report, covering the period October 1969 – September 1970. The second Report is preserved (October 1970 – September 1971), then there is another hole for 1971-1972. From then on the series is complete. The next in line (October 1972 – September 1973) is labelled as being the fifth (it should actually be the fourth). Since 1978 the period covered by the Reports was made to coincide with the solar year.



entrance of Britain in the CECAM membership, British scientists took an active part in CECAM's activities from the beginning.<sup>20</sup> As for Germany, on the other hand, CECAM only attracted a few German theoretical chemists who were in contact with Moser. Moreover DFG (Deutsche Forschung Gesellschaft, the German national research council), was not authorized to grant international funding: this was a German application of the theory of the separation of duties. DFG was solely concerned with research inside Germany, whereas the policy of international research was the exclusive responsibility of Ministries, a circumstance that prevented DFG to participate to an enterprise such as CECAM.

### Widening scopes

“Those of you who will have read the first report will note that there is an increase not only in our activity but also in the subjects which have been dealt with in our Centre”; this is the opening sentence of Carl Moser's introduction to the second report of the scientific activity of CECAM, covering the period from October 1970 to September 1971. Unfortunately, the first report not being available in the surviving files of CECAM's early period, it is impossible to judge exactly to what extent the range of scientific subjects covered by the centre's activities had increased from the previous year; for sure, however, already in its second year of operation CECAM had hosted several scientists whose interests were not confined strictly to quantum chemistry. Besides support given to individual visitors, two colloquia had been organized on models for X-ray, neutron and electron diffraction, and on the calculation of the electronic structure of solids. Even more interesting, however, was the invention of a new way to organize and conduct a research project. In 1970 had already appeared the tool that was going to establish itself as the trademark of CECAM's style for cooperative research: the workshop, in the very specific sense that this word acquired at Orsay.

“This report includes the first results of our interest in direct methods in crystallography, and these have in large part been obtained from a six-week workshop. We have already organized, and plan to continue to do so in the future, the usual 3-5 days colloquia. The workshop on the contrary means getting together a small number of scientists (the optimum will depend on the

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<sup>20</sup> “The UK came into CECAM late when Malcolm Haines and myself wrote a memo to SERC in 1979 [sic: actually, in 1979 SERC was still SRC] pointing out the advantages of membership. However, it is characteristic of the flexible way that Carl Moser organises CECAM activities that in spite of our late membership, the UK community were able to take part in CECAM activities from the beginning.” P.G. Burke, *Electron-atom and electron-molecule collision calculations*, in *Perspectives for Computational Sciences in the 1990's*, CECAM, Orsay 1990, p. 5. See also S.J. Smith, B.T. Sutcliffe, *The Development of Computational Chemistry in the United Kingdom*, *Reviews in Computational Chemistry* 10, 1997, pp. 271-316.

subject matter) who are willing to work together for several weeks on one central theme though each one should be encouraged to try out his ideas. The results of the first workshop have been to my mind sufficiently positive so as to encourage us to organize others." No full report of this first CECAM workshop on "Direct methods in X-Ray crystallography" has survived; for sure, among the participants was Herbert Hauptman, who had laid the foundations of the method and would be awarded the Nobel Prize for chemistry in 1985 for its application to a wide variety of chemical structures.<sup>21</sup>

The experience of the workshop was repeated in 1972, when two such events were organized. Most significantly for the future development of CECAM's activities, one of them opened a completely new field, that would become in later years one of the strong points of the science developed at Orsay. In a presentation of the new centre appeared on the CNRS newsletter in April 1972, Moser anticipated that "a suggestion by a Dutch colleague has led the Scientific Committee to take the decision to organize a new workshop dedicated to Monte Carlo calculations and molecular dynamics of water, to be held in summer 1972".<sup>22</sup> The Dutch colleague was Herman Berendsen, professor of Biophysical Chemistry at the University of Groningen, and one of the pioneers of molecular dynamics in Europe. The ground-breaking paper by Aneesur Rahman and Frank Stillinger on the molecular dynamics of liquid water had just been published the previous year, and Berendsen's proposal to devote a full workshop to the matter couldn't have come at a more appropriate time. Over a period of almost two months, the CECAM workshop collected on the hill at Orsay a good sample of the best experts in molecular simulation (including Kurt Singer and Ian McDonald from Britain), was the starting point for the durable and fruitful relation between Rahman and CECAM, and opened a full new line of development for future CECAM activities, in a way that was just hinted at in its summary report: "This workshop is the first scientific activity of CECAM devoted to computational physics that is essentially classical mechanics. Previous efforts have all been directed to the application of quantum mechanics to problems in atomic and molecular physics. We plan that

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<sup>21</sup> In a report submitted to CNRS Moser made explicit reference to this case to underline the significance of CECAM's workshops: "A recent event, the attribution of the 1985 Nobel Prize for Chemistry to Herb Hauptman, shows how influent CECAM's workshops can be. Though the early papers by Hauptman and Carle date back to the mid-fifties, the CECAM workshops in 1970, 1973 and afterwards have largely contributed to make the community of experts in crystallography aware of the potential power of the so-called direct methods. A common interest could develop this way, to a point that would have been unattainable without this workshop programme; moreover, Hauptman has been an active participant to the workshops on direct methods." Report by Carl Moser for the Direction des Relations et de la Coopération Internationales, May 20, 1986, CNRS Archives, Dépôt de Gif-sur-Yvette, versement 910001.

<sup>22</sup> C. Moser, *Le Centre Européen de Calcul Atomique et Moléculaire*, Courier du CNRS, April 1972, pp. 26-27.

this workshop is only the first of many where a particular interest will be the applications of molecular dynamics to problems of physics and chemistry and eventually other types of problems where classical mechanics may be of interest".<sup>23</sup> Molecular simulation had come to CECAM, and was there to stay.<sup>24</sup>

Soon a new working tool was tested. The Council agreed that the selection of the topics around which to organize future research activities would have benefited from preliminary debate about the relevance, and the perspectives for future advancements, of the subject matters proposed for the workshops to come. The novel format was announced in the Scientific Report for 1972-1973: "We believe the organisation of workshops is one of the most original features of our Centre and that if these are to be most successful, they should be carefully prepared, if possible, at small preliminary meetings". Two such "discussion meetings", as they came to be usually called, were indeed held in 1973 and led to the organisation of three workshops the following year. "Small" meant in fact that these meetings were supposed to last only a few days, contrary to the long duration of the workshops (between one and two months). By that time, the pattern that would be followed in the future was set: each year CECAM would organize a number of workshops, putting together a limited number of people working on a well-defined subject for an extended period of time and actually doing research making full use of CIRCÉ's computing facilities, and a few short meetings of larger groups of interested scientists to discuss the state of the art in a given research sector, evaluate the potentialities for a successful cooperative effort on that specific field, and submit proposals for further workshops, to be in the end approved or rejected by the Scientific Council.

It is not the aim of this paper to provide a detailed and exhaustive account of CECAM' scientific activity and results. A reasoned and critical survey has been made by Herman Berendsen in his contribution to the booklet produced

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<sup>23</sup> *Molecular Dynamics and Monte Carlo Calculations on Water*, Report of a CECAM Workshop, held in Orsay, June 19 – Aug. 11, 1972, CECAM Archives, Lausanne.

<sup>24</sup> Indeed, molecular simulation had already landed, rooted and developed in Orsay, even before CECAM settled there. At the Theoretical Physics Laboratory on the lower part of the Orsay campus, Loup Verlet and his group had given pioneering contributions to the field since 1967, making use of the Univac computers of the Orsay University Computing Centre, and maintained close ties with several of the same experts which Moser hosted at his Centre up on the hill. For various reasons, in spite of their physical proximity and community of interests, the Verlet group and CECAM "superbly ignored each other" for quite some time. This rather peculiar story is briefly recalled by Jean-Pierre Hansen and Dominique Levesque in their contribution to *Recollections of CECAM – For Carl*, CECAM, Orsay 1990, p. 31. A detailed account of Verlet's contributions to the field is given by the same authors: D. Levesque, J.P. Hansen, *The origin of computational Statistical Mechanics in France*, European Physics Journal H 44, 37-46 (2019).

in 1990 on the occasion of Moser's retirement.<sup>25</sup> Berendsen gives a full list of the workshops and discussion meetings held over a period of twenty years, from CECAM's birth to 1988, arranged in six tables corresponding to six different (though often related) main areas into which he splits the research activity developed at CECAM. These are 1) Direct methods in X-ray diffraction, 2) Plasma's and laser fusion, 3) Quantum mechanics in chemistry and solid state physics, including the study of excited states, 4) Molecular dynamics, 5) Biological applications, 6) Reaction paths and rates. Boundaries between these areas were often not rigid, and indeed fruitful cross-fertilization across different fields was one of the main results of successful workshops. Possibly the best example was given by the celebrated 1976 workshop on "Models for Protein Dynamics", where, in addition to the important results obtained (during the workshop the first protein simulation was carried out, by Andy McCammon), a significant accomplishment was to put effectively at work together for the first time scientists working on molecular dynamics and biological matters: "the accurate simulators and the crude biophysicist formed two groups with highly deviating interests and preferences. But there was a lot of interaction and some could bridge the gap".<sup>26</sup>

The Scientific Report for 1976-1977 provides useful information allowing to trace a summary balance of CECAM's first eight years of activity. In the annual 1976 meeting of the Scientific Council it had been decided to hold in September of the following year a conference to celebrate the tenth anniversary of the Blaricum meeting, where the foundations of CECAM had been laid.<sup>27</sup> In the end, the conference did not take place, but the recurrence was nonetheless duly noted the Scientific Report: "The year of 1967 was the tenth anniversary of the meeting in March 1976 (sic!), which was held in Blaricum, Holland and which led to the organization of CECAM. It seemed to us worthwhile to take stock of the activity of our Centre by compiling a list of those scientists who have actively participated in our Centre's research activities and the publications which have resulted from their stay in Orsay."<sup>28</sup> To sum it up in a few numbers, in eight years 18 workshops and 20 discussion meetings had been organized; 306 scientists from 22 different countries (14 of them European) had spent some time as visitors of the Centre, while 198 papers, more or less directly connected with research conducted at CECAM,

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<sup>25</sup> H.J.C. Berendsen, in *Recollections of CECAM – For Carl*, CECAM, Orsay 1990, pp. 2-18.

<sup>26</sup> H.J.C. Berendsen, *The Development of Molecular Dynamics at CECAM*, in J.P. Hansen, G. Ciccotti, H.J.C. Berendsen (eds.), *In Memoriam Aneesur Rahman 1927-1987*, CECAM, Orsay 1987, pp. 9-14. A measure of the historical relevance of this workshop is given by the fact that a conference was organized to celebrate its 40<sup>th</sup> anniversary: *Models for Protein Dynamics. 1976-2016*, held at CECAM Headquarters in Lausanne on February 15-18, 2019.

<sup>27</sup> Compte-rendu du Comité Scientifique du CECAM – 27/11/76 à Orsay, CECAM Archives, Lausanne, Box 050.

<sup>28</sup> Rapport d'activité scientifique du CECAM, 1er Octobre 1976 au 31 Décembre 1977, CECAM Archives, Lausanne, p. 4.

had been published in the scientific literature. Clearly, the impressive list of over three-hundred names (which anyhow does not include scientists who attended only discussion meetings) does not allow to distinguish between those who just paid a short visit to Orsay, and those who went there repeatedly and were actually residents for an extended period of time. Nor can be exactly told which of the listed papers were actually written during CECAM stays, thanks to the availability of the computing facilities there. Most of all, what impresses is the extremely wide range of countries represented among visiting scientists. Already in its early years of operation CECAM had succeeded in establishing itself as a genuine international pole of attraction for researchers working in fields that were no longer restricted to quantum chemistry, but covered an expanding and broad spectrum of interests. And the attraction worked for the younger people at the beginning of their career and for the senior scientists; both found at CECAM working facilities often unavailable at home and a stimulating intellectual environment. Daan Frenkel gives a lively description of the “atmosphere of structured chaos” reigning in Orsay when he first arrived there in May 1975 from Amsterdam as a young PhD student: “The atmosphere at CECAM was truly international: a constant flux of Gods and semi-Gods in science, people whose names I had thus far only read in awe, appeared in person, gave seminar, stayed for dinner, and by large changed perception of science”.<sup>29</sup> And he goes on with an appreciation of the “style” that allowed that atmosphere to survive and provide the proper environment for intellectual creativity: “I consider it the lasting merit of Carl Moser that he has managed to keep this spirit alive in this time of “business-like” science management, where the average administrator wants the scientific discoveries of the next five years to be written down in advance and in triplicate. Carl never joined the paper-shuffling game. The impact of CECAM can therefore not be gauged by reading the annual CECAM reports. Unless you simply look at the list of names, and count the number of European scientists who, effectively, started their computational work at CECAM. Then the picture becomes very clear”.

## **Into the 80's**

While CECAM was performing successfully on scientific grounds, thanks also to his director's pragmatic behaviour and disregard for the “paper-shuffling game”, the persisting lack of a clear definition of its legal status continued to be a source of uneasiness for the top CNRS officials (and, to some extent, for the partner institutions). The “more definitive solution” that Curien was invoking in 1970 had not seen the light over ten years later. To maintain a high level of autonomy and operational flexibility, Moser refrained from creating rigid bureaucratic norms, while the peculiar character of his centre made it hard for

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<sup>29</sup> D. Frenkel, in *Recollections of CECAM – For Carl*, CECAM, Orsay 1990, p. 30.

CNRS to find a satisfactory solution to its institutional status. Was CECAM a CNRS structure, or an independent body? On one hand, from the institutional point of view, CECAM didn't even exist: the visitor entering building 506 in Orsay wouldn't find a single indication that the offices of something called CECAM were located there on the sixth floor.<sup>30</sup> On the other hand, the heading of the official correspondence sent from those offices read: Ministère de l'Éducation Nationale/Centre National de la Recherche Scientifique/Centre Européen de Calcul Atomique et Moléculaire, showing a hierarchical sequence that presented CECAM as a subordinate body to CNRS. Even on the official birthdate of the Centre there was no agreement; while Moser declared it to be October 1969, CNRS documents postponed the event to 1970. A different perception of the true nature of the Centre was developing: Paris administrators regarded CECAM as a CNRS laboratory open to European collaboration, while the "Moser family" thought of it as an independent European organization that happened to be hosted by a CNRS facility.

Besides the formal side of the question, CNRS felt that its own share of participation to the overall budget of CECAM was growing out of proportion compared to the real benefits for France. The amount of the yearly contribution of each of the member institutions was of the order of 2- to 300.000 French francs; CNRS gave about the same in terms of direct funding, but was burdened with further contributions given by salaries of director and staff, lodging, and, most heavily, computing hours at CIRCÉ that were offered to CECAM users at a very convenient discounted price. At the end of 1979 the CNRS representatives in the Scientific Council estimated that the real financial participation of CNRS to CECAM expenses had been "in ten years oscillating between 50% and 85%".<sup>31</sup> Foreign partners, on the other hand, were at times complaining about the way in which the overall budget of CECAM was administered, and wished for a tighter control of the Council over the Centre's financial management.

The issue was explicitly raised when, at the end of 1976, the convention between CNR and CNRS defining the Italian participation to CECAM had to be renovated. Apparently, CNR proposed to modify the previous rules and establish that Italian funds would only be used to pay for the visits to Orsay of Italian scientists. Moser's reaction was that "if that same rule applied to all partners, there would be no CECAM". He insisted on the policy that "an international organization, even a small one like CECAM, can only work efficiently if there are common research programmes and a common financing

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<sup>30</sup> "CECAM occupies a part of the building which houses the Computing Center of the French National Science Foundation (CNRS). The building has a number and the name of the Computing Center (CIRCE) but CECAM' name does not appear"; C. Moser to J.B.H. Otker (Z.W.O.), September 16, 1985, CECAM Archives, Lausanne, Box 043.

<sup>31</sup> Compte-rendu de la réunion du Comité Scientifique du CECAM, 30 novembre 1979, CECAM Archives, Lausanne, Box 050.

of these projects".<sup>32</sup> Writing on that matter to CNR's President Ernesto Quagliariello he made this point clear: "It has never been contemplated that each one pays for his own scientists. I'm convinced that up to now the present formula has been beneficial for everybody, and most of all for Italian scientists".<sup>33</sup> And then he played his winning card: "If we propose that each partner covers only the expenses of his own researchers, this should apply to all partners in our Centre, including French bodies such as CNRS and CEA. One should therefore foresee that the cost of the computations made at CECAM would be supported by each body, according to the use made by his own scientists. In that case, the amount of your contribution to CECAM for 1977 would be much greater than the present estimate of 50 million lire!" An agreement was in the end reached, and the new convention with CNR signed, only in October 1978, for a duration of three years.<sup>34</sup>

A report sent in March 1981 to CNRS Director General Jacques Ducuing summed up the situation and the pending problems regarding the functioning of CECAM.<sup>35</sup> It stated that, "created in 1970, CECAM has never been given a legal structure"; that an RCP (Recherche Coopérative sur Programme) du CNRS had been created to that end in 1978, but had just been abandoned; that a structure of the kind GIP (Groupement d'Interet Scientifique) had been envisaged as a possible way to solve the problems related to CECAM administration. For the financial side of the matter, the report underlined the amount of the double (direct and indirect) contribution by CNRS. The provisional financial balance of 1980 gave an overall expense of about 1.500.000 FF (covered by the direct contributions of the partner institutions); about half of that sum was what CECAM paid to CIRCÉ for computing time. This was actually a limited portion of the real cost of the computing time used by CECAM (it used to be around 20% in the early years and had grown up to 60% by 1980). The estimated "indirect" CNRS contribution to the real total CECAM expenses amounted to about 500.000 FF.

The report went on presenting the preoccupations of (some of) the

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<sup>32</sup> C. Moser to Mme Mirabel, CNRS Relations Internationales, 2 décembre 1976. CNRS Archives, Dépôt de Gif-sur-Yvette, versement 910001.

<sup>33</sup> C. Moser to E. Quagliariello, 2 décembre 1976. CNRS Archives, Dépôt de Gif-sur-Yvette, versement 910001. Moser had a point in claiming that Italy had possibly benefited from CECAM more than any other partner country. This was, at least, the feeling of visitors to Orsay. According to Daan Frenkel and Jean-Paul Ryckaert, CECAM in the mid-seventies was "nothing else than an Italian enclave", and "an Italian colony": see their contributions in *Recollections of CECAM - For Carl*, CECAM, Orsay 1990, pp. 30 and 34 respectively.

<sup>34</sup> At the end of 1981, however, due mainly to internal tensions about its overall scientific policy, CNR withdrew its participation as a partner member, until in 1986 the Convention defining the CECAM status was approved. Nonetheless, between 1982 and 1986 Italian scientists took part in CECAM activities, their expenses being directly covered by their institutions.

<sup>35</sup> W. Mercouroff (Direction des Relations Exterieures), Note à l'attention de Monsieur Ducuing, Paris, 20 mai 1981. CNRS Archives, Dépôt de Gif-sur-Yvette, versement 910001.

foreign partners: "Since several years, the main CECAM foreign partners, namely M. van Lieshout, Director of ZWO, and the representatives of the Italian CNR have communicated to CNRS that they consider CECAM's activity as being of particular interest, but that they are not satisfied of the way it is administered... The foreign partners wish that M. Moser could no longer make uncontrolled handling of the credits at his disposal, and think that a well-established structure would allow to solve these difficulties".

The note that Ducuing sent privately to Mercouroff upon receiving the report leaves no doubt about the feelings prevailing among the upper echelons of CNRS: in the benefit-to-cost competition between the several partners of the collaboration, the French scientific community felt distinctly to be by and large the most disadvantaged, to the point that CNRS Director was considering the idea of "putting an end to that all", unless the situation be substantially redressed.<sup>36</sup>

Five years later, a similar report (indeed essentially the same, with some minor modifications required by the few novelties occurred in the meantime, such as the entrance of Great Britain in the group of the foreign partners) was sent to the CNRS Financial Controller, with an urgent request to evaluate the attached proposal for the creation of a Groupement Scientifique.<sup>37</sup> This was the final outcome of a project that "had been elaborated in several occasions with our foreign partners since 1982". In fact, over a period of five years a dozen different versions of that proposal had circulated among the various actors, each time introducing slight modifications to reach an agreement that would satisfy all demands. The definitive version of the convention was signed on October 24, 1986 at the CNRS Chateau, Gif-sur-Yvette. It formalized the creation, by the institutions signing the agreement, of a Groupement Scientifique, the "Centre Européen de Calcul Atomique et Moléculaire", with seat at CIRCÉ in Orsay, having as its aim "to promote the exchanges and contacts inside the international scientific community, to promote scientific collaboration between the main research bodies at the European level, and to set up and coordinate seminars and workshops in those fields where the methods of numerical simulation play a dominant role". The agreement was signed by representatives of CNRS (France), CEA-DAM (France), CNR (Italy),

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<sup>36</sup> "L'interet pour le CNRS et la communauté scientifique française du CECAM n'apparaît toujours pas nettement. Aussi suis-je, actuellement, défavorable à la création d'une structure internationale impliquant le CNRS. J'avoue d'ailleurs ne pas très bien voir la justification du cadeau de 500 KF en heures de calcul. Pour te livrer le fond de ma pensée, je mettrais bien un terme à tout cela, à moins qu'on ne me montre un bénéficiaire français." Note by J. Ducuing to W. Mercouroff, April 16, 1981, CNRS Archives, Dépôt de Gif-sur-Yvette, versement 910001.

<sup>37</sup> J.F. Miquel (Direction des Relations et de la Coopération Internationales), Note à l'attention de Monsieur J.F. Heyman, Contrôleur financier, Paris, 25 avril 1986. CNRS Archives, Dépôt de Gif-sur-Yvette, versement 910001.



ZWO (Holland), SERC (Great Britain) and FNRS (Belgium). It was meant to be valid for a period of three years, and it was intended to be renewed by tacit agreement unless explicitly denounced by one or more of the member parties.

In its essence, the convention just limited itself to formalize the existent situation, and not much changed afterwards in the usual functioning of CECAM; however, by putting explicitly on an official document the rules of the game, defining the composition and the functions of the Scientific Council, the tasks and prerogatives of the Director, fixing the annual contributions to be versed by the member organizations, the convention finally gave CECAM the legal structure it had been lacking since its foundation. The main innovation concerned the Director: he (or she) was to be designated by the Scientific Committee, and would stay in charge for three years, with a possibility for a second mandate. This condition wiped out any problem potentially arising with CECAM's founding father and eternal Director, Carl Moser, who would in any case be forced to retirement in 1990. At the first meeting of the Scientific Council of the "new" CECAM, held on March 20, 1987, Moser was confirmed in his function of Director, for three years starting January 1, 1987, "under reserve that it be consented by the law regulations on the age of retirement".<sup>38</sup>

## Continuity and changes

In 1990 two CECAM publications, of quite different character but in some sense related, saw the light. One was a collection of personal reminiscences by some of the protagonists of the Centre's life during the "Moser kingdom", prepared as a tribute to Carl Moser on the occasion of his retirement.<sup>39</sup> Daan Frenkel had circulated the proposal of producing such a collective memory during a special meeting held at Orsay in October 1989 to celebrate the twentieth anniversary of CECAM. The proceedings of that meeting were also published in 1990.<sup>40</sup> Though the meeting was intended to provide insights for the future, looking forward to potential developments, the list of the subjects covered give a fair representation of the impressive expansion in the range and scope of the scientific fields covered by CECAM's activities since its creation as a centre solely oriented to quantum chemistry. And some of the authors, before discussing future possibilities, introduced their talks by a retrospective look at the way in which CECAM had contributed to the development of their specific field.

In fact, all along the 80's CECAM's scientific activity had kept expanding, both in the number of initiatives and in the range of subjects covered. Here

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<sup>38</sup> Compte-rendu de la 1<sup>ère</sup> reunion du Conseil CECAM qui s'est tenue au siège du CNRS à Paris le mardi 20 Janvier à 14h30. CNRS Archives, Dépôt de Gif-sur-Yvette, versement 910001.

<sup>39</sup> *Recollections of CECAM – For Carl*, CECAM, Orsay 1990.

<sup>40</sup> *Perspectives for Computational Sciences in the 1990's*, CECAM, Orsay, 4-6 October 1989.

again, for an accurate evaluation the reader is referred to the reconstruction provided by Berendsen. A few numbers give an idea of that growth: while in the decade 1970-1979 CECAM had organized 30 workshops and as many discussion meetings, these numbers grew in the following decade (1980-1989) respectively to 80 workshops and 72 discussion meetings (that sometimes also took the name of preliminary, or planning, meeting). Such had been the growth of the different subjects covered, far away from the original confinement to computational chemistry, that Moser was led to comment that "There are some who feel that the program of CECAM workshops doesn't have nearly enough "chemistry" subjects. We seem to be concerned only with physics, astrophysics and molecular biology".<sup>41</sup>

There were, however, a few novel problems arising along with this enlargement of activities. One was a crude problem of space. If in 1969 CECAM could be happily hosted in Building 506 thanks to the amount of free space there, by the late eighties the increasing need of more room for the expanding computing facilities of CIRCÉ came into conflict with the growing number of workshops and related activities, requiring seminar rooms that were no longer easily available. Moser had to raise the issue with CNRS Scientific Director and ask for his intervention to define "the rules of "cohabitation" between CIRCÉ and CECAM in this building" when the director of CIRCÉ planned to appropriate one of the two CECAM seminar rooms and convert it into offices for his group.<sup>42</sup> Cohabitation, somehow, continued.

More relevant on scientific grounds, a significant change intervened throughout the 80's in the organization of the workshops. While their number grew steadily, their duration was drastically shrinking. On the average, the 30 workshops held in the first decade had kept the participants occupied for a period of seven weeks. Between 1980 and 1989, the 80 workshops lasted on average only two weeks; some of them, more and more as time passed by, only about a week. There were several reasons for that. Besides organizational problems (holding more meetings necessarily meant less time to devote to each one of them on CECAM premises, which also explains why a growing number of workshops did not take place at Orsay), the increasing diffusion of computing facilities throughout Europe made it unnecessary to stay for long periods of time at Orsay to exploit the once unique possibilities offered by CIRCÉ. People could now envisage workshops being just an occasion for theoretical discussions and elaboration of projects, while the actual computational work could be more easily performed at their domestic institutions. The organizers of the workshops had to find the best compromise to lighten the burden of heavy computational costs (in terms of time and money) while keeping the meetings long enough to guarantee effective intellectual

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<sup>41</sup> C. Moser to W. Niewpoort, January 20, 1986. CECAM Archives, Lausanne, Box 043.

<sup>42</sup> C. Moser to J.C. Lehmann, July 15, 1987, CECAM Archives, Lausanne, Box 048.

interaction between participants, a characteristic of CECAM workshops regarded by many as the fundamental key to their success, even more than the availability of excellent computing resources. CECAM's "old guard" insisted on keeping workshops as long as possible, and both Moser and his successor Giovanni Ciccotti encouraged the renewal of the former policy of having at Orsay long-time visitors, an habit that had been slowly disappearing.

Computer development was a subject very dear to Moser, who devoted a good part of his personal efforts to that matter. A discussion meeting on "Special Purpose Computational Machines" was held in Holland in 1984. Soon after, in close collaboration with Berendsen, Moser tried for a while, relying on his extended network of personal relations, to set up a joint effort for the creation of a European centre for the construction of special purpose parallel architecture computers. A thick correspondence on the subject is preserved in Moser's files, involving not only scientists of the CECAM inner circle, but the wider community of people working at the time to design new computer architectures tailored for specific scientific tasks. In the end, mainly because of the lack of the necessary industrial support, nothing came out of that proposal; the issue of future developments of computer design, however, remained one of Moser's main concerns.

Among the different research areas investigated by individual visitors and discussed in workshops, molecular simulation may serve as an example to judge the relevance of CECAM related contributions to the advancement of the field. Already in the 70's, following the 1972 workshop on the simulation of water, a workshop held two years later on "Methods in molecular dynamics – Long time-scale events" was the starting point for the introduction of the mass tensor technique by Charles Bennett, while at the same time, during the long periods of time they spent at Orsay, Gianni Jacucci and Giovanni Ciccotti developed their subtraction technique, and soon after Ciccotti and Jean-Paul Ryckaert started the work on constrained dynamics that incorporated the SHAKE algorithm just in time for it to be used during the 1976 workshop on the simulation of proteins. Moving into the 80's, it was during a 1983 workshop that Daan Frenkel and Tony Ladd wrote their paper on the calculation of free energies; in 1984, the encounter between Shuichi Nosé and Bill Hoover at another CECAM molecular dynamics workshop led Hoover to work on his modified version of the Nosé thermostat, while a group of participants, including Nosé and Hans Christian Andersen, produced a detailed report on "New molecular dynamics methods for various ensembles". In the summer 1985, CECAM organized at Orsay the first workshop devoted to a stochastic approach to chemical reactions, a topic that was resumed in three following summer workshops, from 1986 to 1988, all of them dedicated to "Computer simulation and the theory of chemical reactions in solution", whose outcome was the development of the "Blue Moon Ensemble". Some of these results were reached during the workshops themselves, others resulted from further

elaboration of ideas originated in workshop discussions. And, beyond the achievements that were direct results of CECAM activities, there is little doubt that in general CECAM acted throughout this period as an effective “resonance box” for ideas that, even if produced elsewhere, found in the peculiar international atmosphere of the workshops the perfect environment to diffuse and be circulated among the scientific community at large.

The issue of the choice of Moser’s successor was discussed by the Council at the 1988 meeting, when the procedure for the appointment of the new director was established. It was agreed that the directorship would be a full-time function, and that the choice should fall on an active scientist, whose administrative duties were to be kept at a minimum, thus allowing him to devote the major part of his time to scientific activities relevant to CECAM.<sup>43</sup> Candidatures were advanced throughout the following year. Following interviews with prospective candidates in June 1990, the final choice of the Council fell on Giovanni Ciccotti, professor of Molecular Physics at the University “La Sapienza” in Rome. As a computational theoretical physicist, Ciccotti was really a “CECAM’s child”: his long-standing activity in molecular dynamics and statistical physics had started during a stay at Orsay in 1974, at the invitation of his friend and colleague Gianni Jacucci, and since then his achievements, both in research and in organizational endeavours, had been strictly connected to CECAM related activities. Ciccotti was officially designed as Moser’s successor on October 1 1990. To give a clear indication of the changing and widening range of scientific areas falling under CECAM related researches, he managed to have them shifted, for the sake of official CNRS evaluation, from section 04 (Atoms and molecules; optics and lasers; hot plasmas) to section 02 (Physical phenomena, theories and models). Ciccotti made a substantial effort to resume the practice of having at CECAM long-term visitors, and, in the course of his mandate, managed to equip CECAM with first-class computer hardware.<sup>44</sup> In the way of managing practices, he definitely shared with his predecessor a nonchalant attitude toward bureaucratic obstacles and a disregard for the “paper shuffling game”, that were difficult to reconcile with the rather stiff handling of such matters by French administrators. These peculiar traits of character would soon be put to test by the new institutional crisis that was preparing.

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<sup>43</sup> CECAM Scientific Council, Meeting of 20 September 1988, “Procedure for Appointment of New Director”, CNRS Archives, Dépôt de Gif-sur-Yvette, versement 910001.

<sup>44</sup> A detailed list of the equipment owned, or on leasing, by CECAM “thanks to Giovanni Ciccotti’s far reaching policy”, is in a report “Computing at CECAM”, undated and with no signature, but most likely compiled by Stefano Baroni and dating from the very early days in Lyon. CECAM Archives, Lausanne, Box 124.

## Leaving Orsay

In 1992, the CNRS Director General François Kourilsky launched MIPS (Moyens informatiques pour la science), a large project aimed at an upgrade and a full renovation of the national resources for computational science. Charged with the implementation of the project was Victor Alessandrini, an Argentinian born theoretical physicist, professor at the Université Paris XI in Orsay, who was since 1988 one of the CNRS representatives in CECAM's Scientific Council. Within the larger MIPS project, the creation of a new supercomputing centre was envisaged, endowed with the best available machines and coupled to a research laboratory, originally designed to be located as a brand-new institution in Marseille. This option revealed itself too ambitious to be realized, and it was decided that the new centre would be installed at Orsay, replacing CIRCÉ that was going to be dismantled. Creating such a supercomputing centre and research laboratory at the Orsay site implied for CNRS reconsidering its relationship with CECAM. At the end of 1992 Kourilsky sent to the President of CECAM's Council an official letter denouncing the convention, effective December 31, 1993. Kourilski suggested that to "optimize our interface with CECAM"... "it seems desirable to us that CECAM becomes the European component of this laboratory, the ensemble of these activities being placed under CNRS responsibility".<sup>45</sup> A more detailed proposal would be soon circulated to SC members.

The CNRS proposal, specifying at which conditions CECAM would be "the European component of this laboratory", was addressed by Alessandrini to the SC members in January 1993. In the accompanying letter, he commented that "we should be able to maintain a reasonable amount of independence in the management of the traditional CECAM activities",<sup>46</sup> but this was not enough to dispel the feeling that in the proposed scheme CECAM would be in some sense a subordinate body to the CNRS laboratory. This perception was manifest in the commenting letter sent to SC members by the Council President, Daan Frenkel: "Although the CNRS plans are, at present, still in a preliminary stage, it was clear from the briefing that we received from Victor Alessandrini that it is envisaged that the present CECAM activities would be incorporated in this novel scientific computing center. This would have consequences for the way in which CECAM operates as the presence of the new super-computer necessitates the CNRS to have more direct control over

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<sup>45</sup> Letter by F. Kourilsky, December 17, 1992, CECAM Archives, Lausanne, Box 122. The proposed CNRS laboratory was called ICI (Institut de Calcul Intensif); it was to be part of the large supercomputing centre, which was actually inaugurated in December 1993 as IDRIS (Institut du développement et des ressources en information scientifique), still its present denomination.

<sup>46</sup> V. Alessandrini to CECAM Council members, January 25, 1992, CECAM Archives, Lausanne, Box 122.

personnel working in the same building. In particular, it was explained that, in the new structure, there would be only one director who would be appointed directly by the CNRS. This director would be in charge of both the national and the European scientific activities. Moreover, long-term visitors would also have to be approved by the CNRS".<sup>47</sup> Frenkel asked the Council members to be informed on their views on the matter, to be discussed at the extraordinary Council meeting scheduled for March 8, and in particular "to consider explicitly how much of the independence of CECAM your organisation is willing to trade in exchange for the possibility to have access to the super-computer(s) at Orsay".

The reactions of the representatives of CEA, CNR, FNRS, NOW and SERC to the CNRS proposal were enclosed in the preparatory material for the March meeting. In the joint SERC-NOW discussion paper it was explicitly stated that the Council faced three options: a) closing CECAM on Dec. 31, 1993, b) moving CECAM to a different site, c) retaining CECAM at the Orsay site, in symbiosis with the ICI. Frenkel made it clear that at the meeting only option c) would be discussed; other options would be considered only if "unexpectedly, no satisfactory arrangement for option c) can be found". Nonetheless, already at least two different informal proposals to host CECAM were advanced before the meeting took place, by Michel Mareschal from the Université Libre de Bruxelles, and by the director of the Italian computing centre, CINECA, near Bologna.<sup>48</sup>

At the March meeting, a consensus was reached that "it should be possible to arrive at a new CECAM convention that would be satisfactory both to the CNRS and to the other partner organizations". A hectic period followed, with several drafts of the new convention being circulated and going back and forth, but no satisfactory solution was found. Another note by Alessandrini further clarified the viewpoint of CNRS: he stated clearly that "maintaining in our Orsay site an organisation with uncorrelated European and international policies, whose main concern would be to drift away from CNRS as much as possible" was an option that could not be considered. He reinforced his point asserting that "CNRS management of CECAM is required by the very strong coupling we intend to set up with the future supercomputing centre", and claiming that "CECAM has always been a CNRS laboratory with an European projection".<sup>49</sup> Clearly, the circumstance that Alessandrini happened to be at the

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<sup>47</sup> D. Frenkel to CECAM Council members, January 29, 1992, CECAM Archives, Lausanne, Box 122.

<sup>48</sup> All the relevant documentation is preserved in Box 122 of the CECAM Archives in Lausanne.

<sup>49</sup> V. Alessandrini to CECAM Council members, April 20, 1993, CECAM Archives, Lausanne, Box 122. Regarding the last statement about the CECAM status, it can be interestingly confronted with what Moser was explicitly stating on that same point: "CECAM is a European organization and is not a laboratory of CNRS": C. Moser to K. Morakuma, July 23, 1987, CECAM Archives, Lausanne, Box 048.

same time the CNRS representative in the Council conducting the negotiations on the CECAM matter, and the designated director of the future CNRS supercomputing centre, did nothing to ease the relations. In May, Frenkel informed the Council members that no consensus had been reached yet on a possible compromise, given the essential differences between the draft conventions prepared by Leech and Bertoni (SERC-CNR) and by Alessandrini, mainly related to the role and management of research to be conducted at CECAM.

The peak of tension was reached in June, when it became clear that a solution of the “stalemate” (as Frenkel evaluated the situation) that would allow CECAM’s permanence at Orsay, was not possible. An irritated long letter by Alessandrini, containing harsh judgements on CECAM’s management, reaffirmed the points on which CNRS had no intention to compromise, stated bluntly that “CECAM, in its present form, comes to an end at the end of this year”, and basically gave an ultimatum to the Council, directly asking the question “whether the organisation you represent is willing to pursue an active cooperation with CNRS at the Orsay site”.<sup>50</sup> Frenkel let Kourilsky know that he was “personally worried by the deteriorating atmosphere of the negotiations”. Whatever the outcome of the issue, he added, any attempt to diminish CECAM’s relevance as a research institution was out of the question: “Let me conclude by stating that I feel that the CECAM collaboration has been extremely successful and is worth continuing. In the 25 years since it was founded, CECAM has developed into an institution with a great international reputation that attracts a constant stream of top scientists in the field of computational science. The research experience at CECAM has been a crucial formative experience for many of the leading computational physicists and chemists in the CECAM member countries. In fact, without CECAM, Europe would never have developed into world leader in the field of computer simulation. It is clearly important that this leading role be maintained and strengthened”.<sup>51</sup>

The final decision to leave Orsay was taken by the Scientific Council at the extraordinary meeting held in September. It was agreed that the conditions allowing to take what seemed to many the most desirable solution, i.e. to maintain CECAM at Orsay, could not be satisfied, and that the remaining alternative option was to move CECAM to a different location. A vote was taken in that sense, and the resolution to move passed with the favourable vote of the representatives of CNR, SERC, NWO and FNRS; CEA abstained.<sup>52</sup> Proposals to host CECAM (due to leave Orsay by the end of the year) rapidly

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<sup>50</sup> V. Alessandrini to CECAM Scientific Council members, June 2, 1993, CECAM Archives, Lausanne, Box 122.

<sup>51</sup> D. Frenkel to F. Kourilsky, June 18, 1993, CECAM Archives, Lausanne, Box 122.

<sup>52</sup> Minutes of the extraordinary meeting of the CECAM Scientific Council held on 9 September 1993, CECAM Archives, Lausanne, Box 122.

came from ten different sites, alongside with requests to enter as member of the CECAM partnership by the Swiss FNS (Fonds National Science), the Greek FORTH (Foundation for Research and Technology – Hellas), and the Université Libre de Bruxelles, the École Normale Supérieure in Lyon, and SISSA (Scuola Internazionale Superiore di Studi Avanzati) from Trieste, these latter in case their proposal to host CECAM be accepted. At the regular annual meeting of the Council, in November, the site of Lyon was selected: the future CECAM would be hosted by the École Normale Supérieure at Lyon. Instrumental in the matter was the role played by Jean-Pierre Hansen, a computational theoretical physicist who had been a member of Loup Verlet's group in Orsay and had developed friendly relations with the "CECAM family"; he had moved in 1987 from Paris to Lyon, as research director, to help establish there the new École Normale Supérieure, and had founded its Physics Laboratory. The convention defining the status of the new CECAM was elaborated by Carlo Maria Bertoni from CNR and Mme Simoen, general secretary of FNRS. CECAM was an independent association between the member institutions, constituted as a laboratory with the aim "to promote cooperation amongst European research organisations and their scientific communities in furthering research involving computationally intensive methods".

The new premises of CECAM in Lyon were officially inaugurated with a ceremony on April 8, 1994. In the morning of that same day, the Scientific Council had held its first meeting in the new location, and selected as CECAM future Director Stefano Baroni, from SISSA. In his inaugural speech, Council President Daan Frenkel gave his personal evaluation of the state of health of the Centre: "In 1993, when it became clear that the old structure of CECAM had to change, we had to ask: is there still a need for CECAM? I think that this question was convincingly answered by the fact that CECAM received truly excellent offers for new premises from all member countries and even from several countries that had not yet joined CECAM". Indeed, the succession of events originated by the 1993 "crisis" turned out to be a positive indicator of the persisting need of CECAM as an aggregation point for European science. The "CECAM family" was further expanding, and seemed to enjoy good health.



## Acknowledgments

This paper was produced following the suggestion, by CECAM Director Ignacio Pagonabarraga, Deputy Director Sara Bonella and former Director Giovanni Ciccotti that it would be desirable to provide the CECAM community with a sketch of the Centre's early life, on occasion of the 50<sup>th</sup> anniversary of its creation. It is essentially confined to the institutional side of CECAM's history, and even so it has by no means any pretention to be exhaustive or definitive; most likely, the reader will find that important issues are missing, incorrect details are given, and debatable interpretations are suggested. While apologizing for such deficiencies of the present work, I can only express the wish that it may serve as a starting point for further, more complete and extended researches. In preparing the text I have profited, besides the very scarce literature on the subject, of the documentation deposited at the CNRS Archives at Gif-sur-Yvette, thanks to the efficient services of Mme Loïse Scherer, and, most of all, of the CECAM papers that have been in recent times the object of an important archival intervention, and are now properly arranged and deposited at the CECAM headquarters in Lausanne.<sup>53</sup> Among the several persons that have helped me in various ways I want to express my warm appreciation to Victor Alessandrini, Carlo Maria Bertoni, Sara Bonella, Daniel Borgis, Monique Butin, Giovanni Ciccotti, Janine Connes, Antonio Di Carlo, Denis Giroud, Jean-Pierre Hansen, Dominique Levesque, Michel Mareschal, Ignacio Pagonabarraga. It goes without saying that, for all the useful information they provided, any incorrect statement or factual error in the final text are my own responsibility. Finally, I wish to thank warmly CECAM Director Ignacio Pagonabarraga for the support given to this work, and the whole CECAM staff for their hospitality and assistance during my visits in Lausanne.

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<sup>53</sup> C. Abt-Müller, *Organisation et valorisation du fonds du Centre Européen de Calcul Atomique et Moléculaire (CECAM)*, Travail de Bachelor réalisé en vue de l'obtention du Bachelor HES, Genève, August 2017.

