Introduction to

Neutron Spin Echo

Spectroscopy

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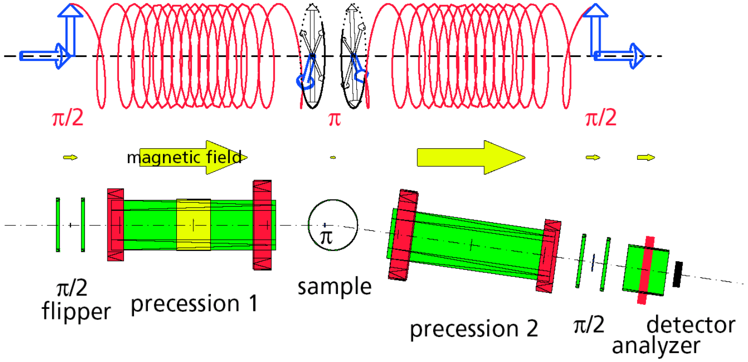
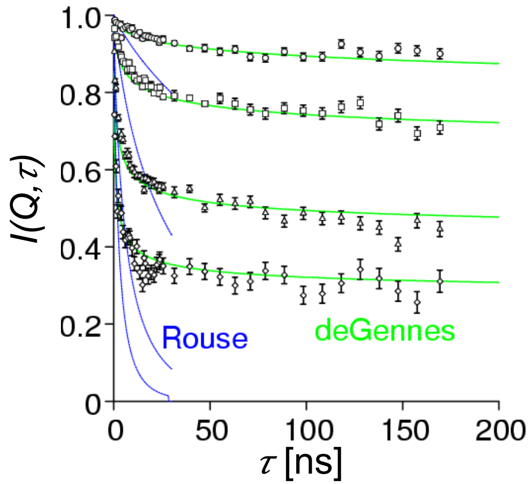
1. Introduction
2. The Principles of NSE
3. The SNS-NSE Spectrometer
4. Examples
5. Summary

Oak Ridge, TN 2017

Neutron Spin Echo in A Nutshell

- NSE measures very small velocity changes by using neutron magnetic moment as a “clock”
- NSE “end-product” is the Intermediate Scattering Function: *I(Q,τ)*
- NSE can use broad wavelength band while maintaining very good resolution
- NSE is a complementary technique to SANS in that it provides the dynamic information about the system
- NSE is “counting intensive”: long counting times, large samples

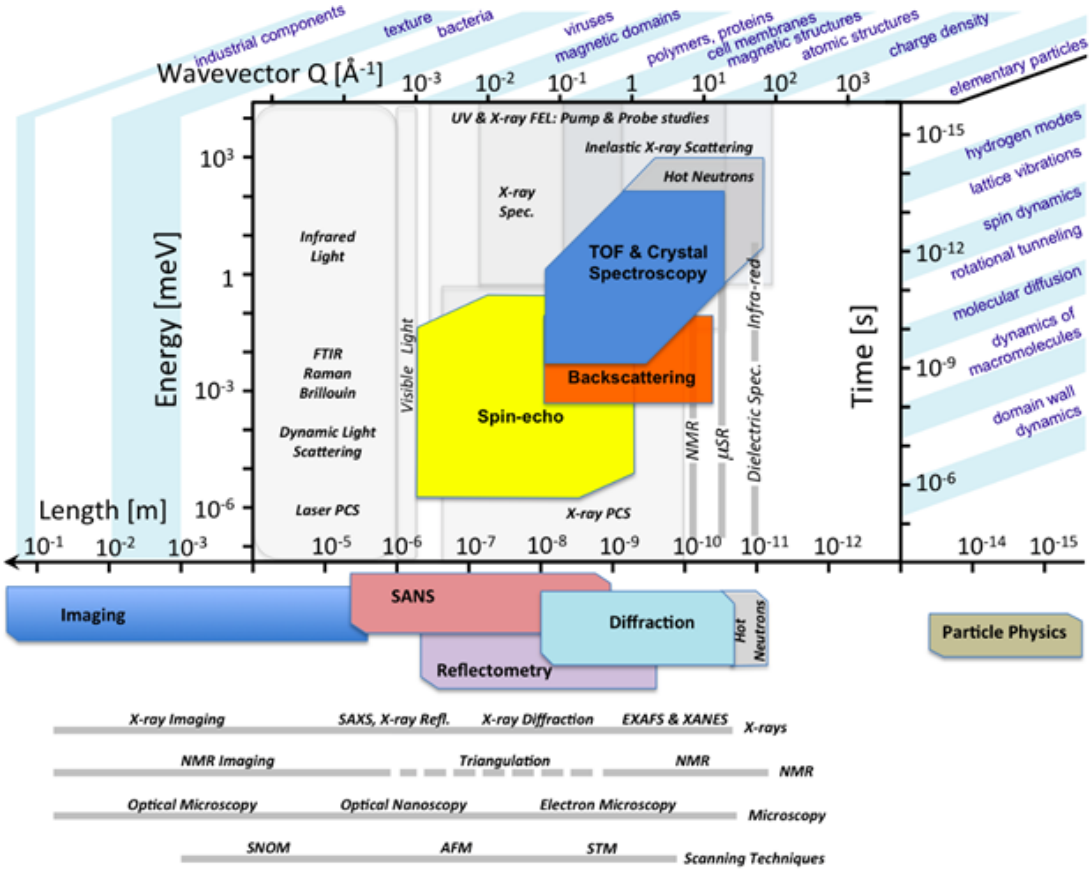




Neutron and X-Ray Instruments Zoology

With NSE one can study:

- Coherent Dynamics
  - Diffusion
  - Shape fluctuations
  - Polymer dynamics
  - Glassy systems
- Incoherent Dynamics
  - Hydrogen
- Magnetic Dynamics
  - Spin Glasses



Source: ESS

NSE Spectrometers in the World

NSE Spectrometers

Classic "IN11-Type"

- IN11 - Institute Laue-Langevin, Grenoble, France
- IN15 - Institute Laue-Langevin, Grenoble, France
- J-NSE – JCNS hosted by FRM-II, Garching, Germany
- NG-A NSE – NIST, Gaithersburg, USA
- C2-3-1 iNSE, ISSP JRR-3M, Tokai, Japan
- BL-15 SNS-NSE – JCNS/ORNL, Oak Ridge, USA

- MUSES [NSRE] – Laboratoire Léon Brillouin, Saclay, France
- RESEDA [NSRE] – FRM-II, Garching, Germany
- VIN-ROSE [NSRE] – J-PARC, Tokai, Japan
- MIRA [TAS+MIEZE] – FRM-II, Garching, Germany
- FLEXX [TAS+NSRE] – HZB, Berlin, Germany
- SESANS [SE+SANS] – TU Delft, Holland

The Principles of Neutron Spin Echo

Inelastic Neutron Scattering

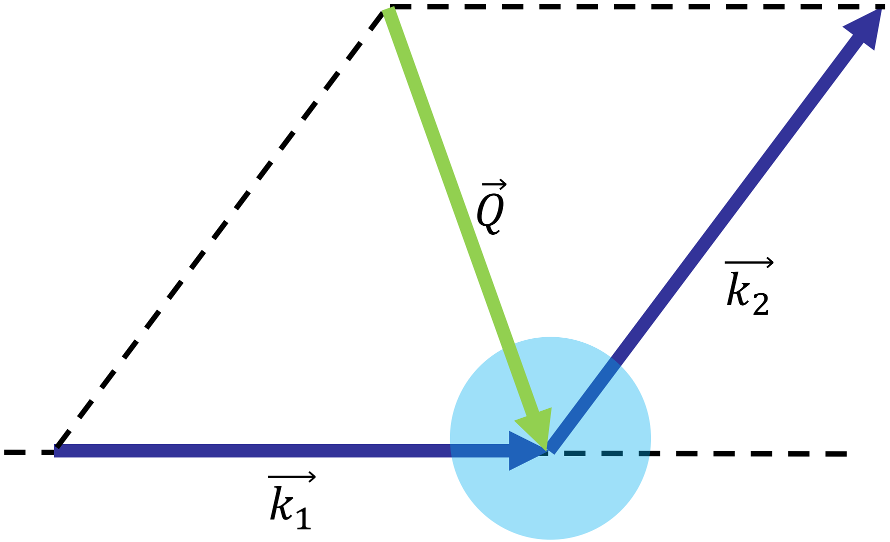
Dynamic Structure Factor

$$\frac{d\sigma}{d\Omega dE} \sim S(Q,\omega)$$

Neutron scattering kinematics:

$$\Delta E = \hbar\omega = (mv_1^2 - mv_2^2)/2$$

$$\vec{Q} = \overrightarrow{k_1} - \overrightarrow{k_2}$$



$$|\vec{k}| = \frac{2\pi}{\lambda}$$

$$\lambda = \frac{h}{mv}$$ (Louis de Broglie, 1924)

Larmor Precession (I)

Bloch Equation:

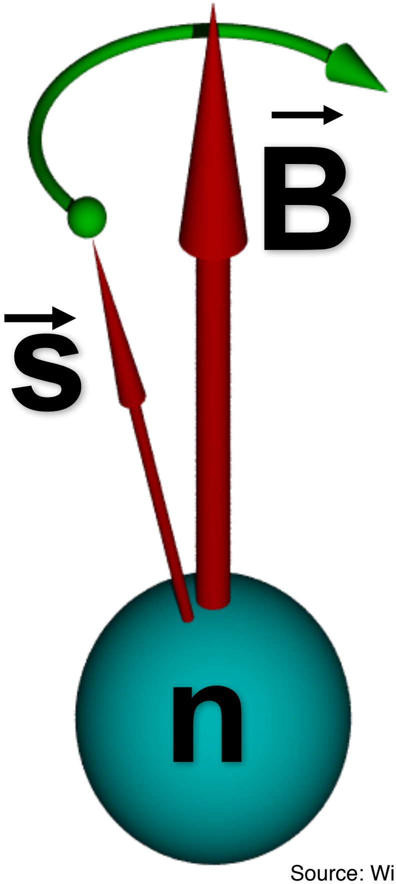
$$\frac{d\vec{s}}{dt} = \gamma \vec{s} \times \vec{B}$$

Larmor Frequency

$$\omega_L = |\gamma B|$$

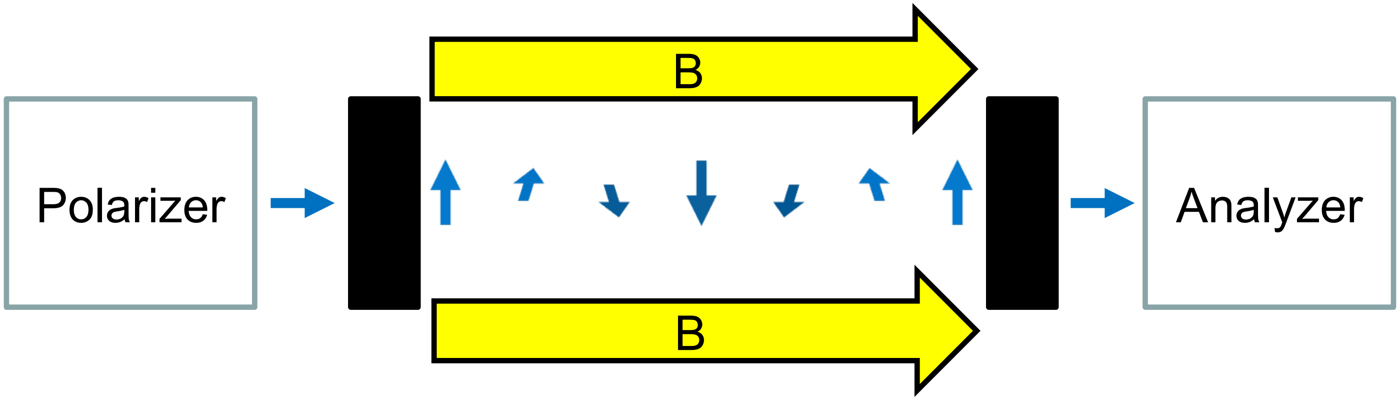
Neutron Gyromagnetic Ratio

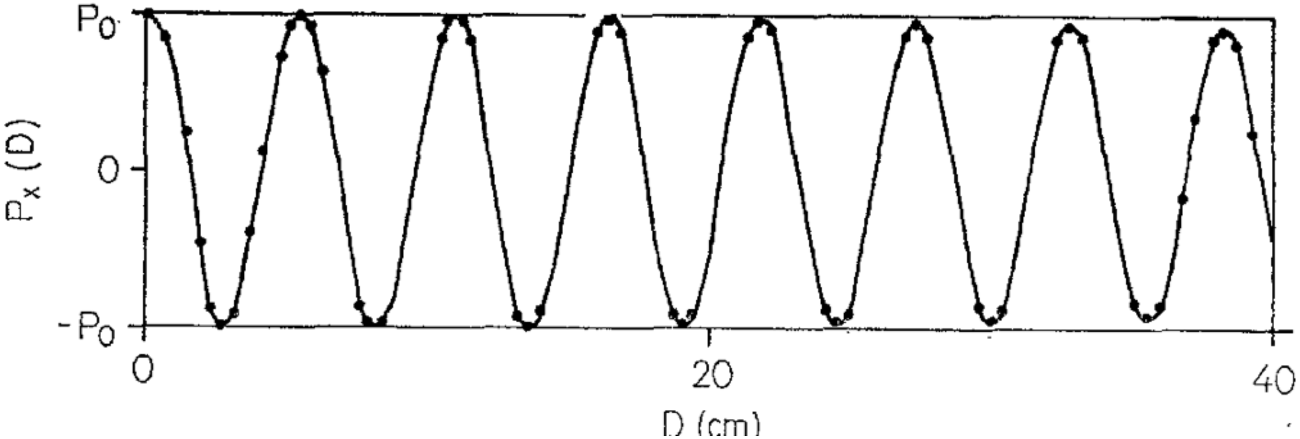
$$\gamma / 2\pi \approx 30\ \mathrm{MHz/T}$$



Source: Wikipedia

Larmor Precession (II)



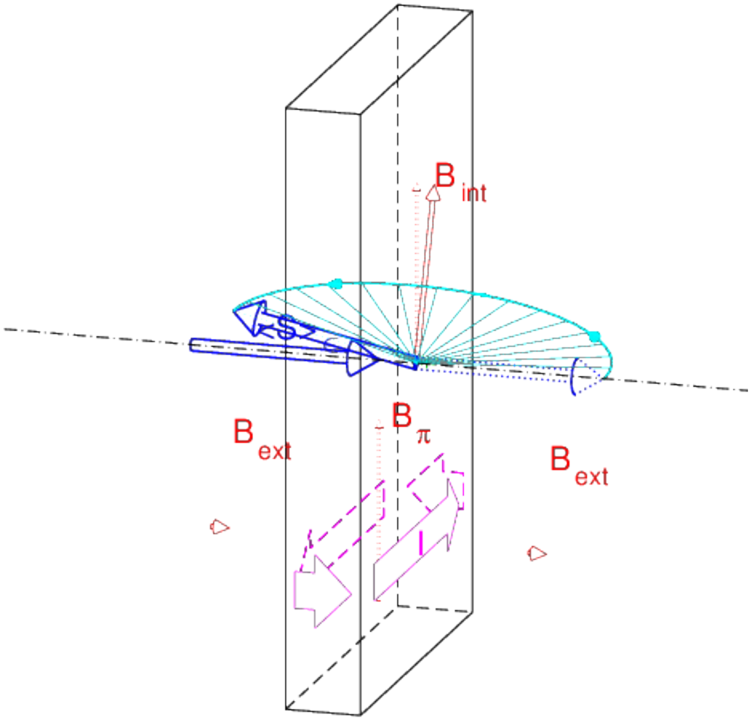


B=1.6 mT λ=1.6 Å

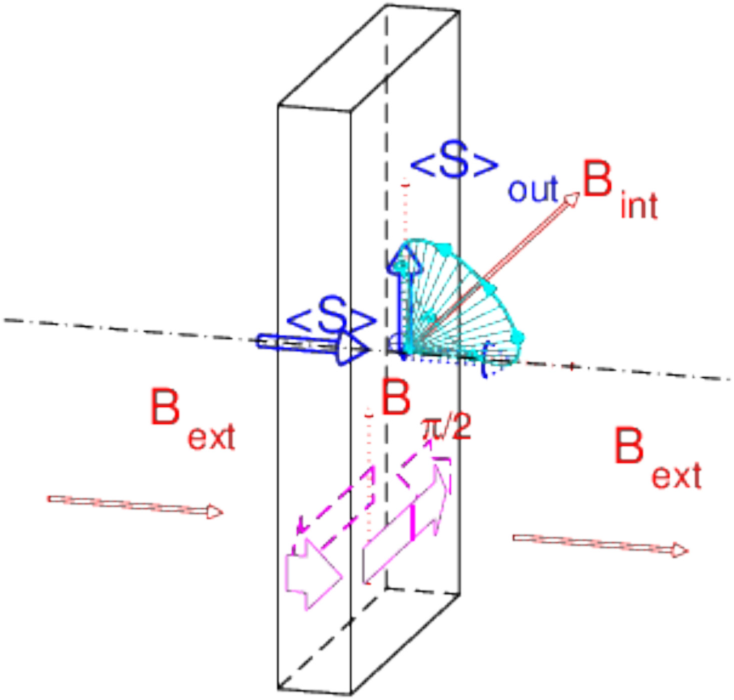
F. Mezei, Z. Phys. 255 (1972), 146-160

Neutron Spin Manipulation
Mezei Flippers

π-flipper



$\pi/2$-flipper

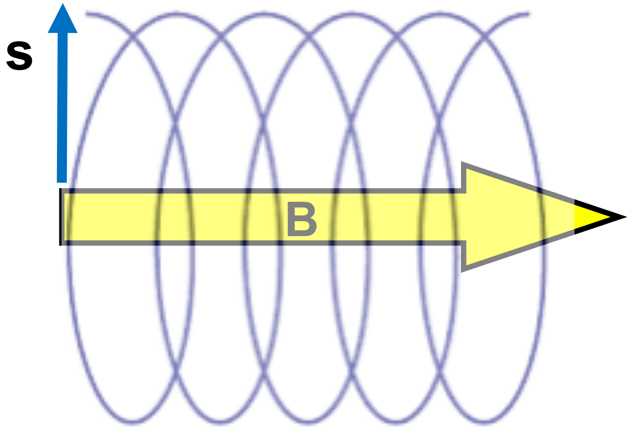


Courtesy: M. Monkenbusch, Forschungszentrum Juelich

Larmor Precession (III)

Accumulated phase:

$$\varphi = \omega_L t = \gamma B l \, \frac{1}{v}$$



or

$$\varphi = \gamma \frac{m}{h} J\lambda$$

for example:

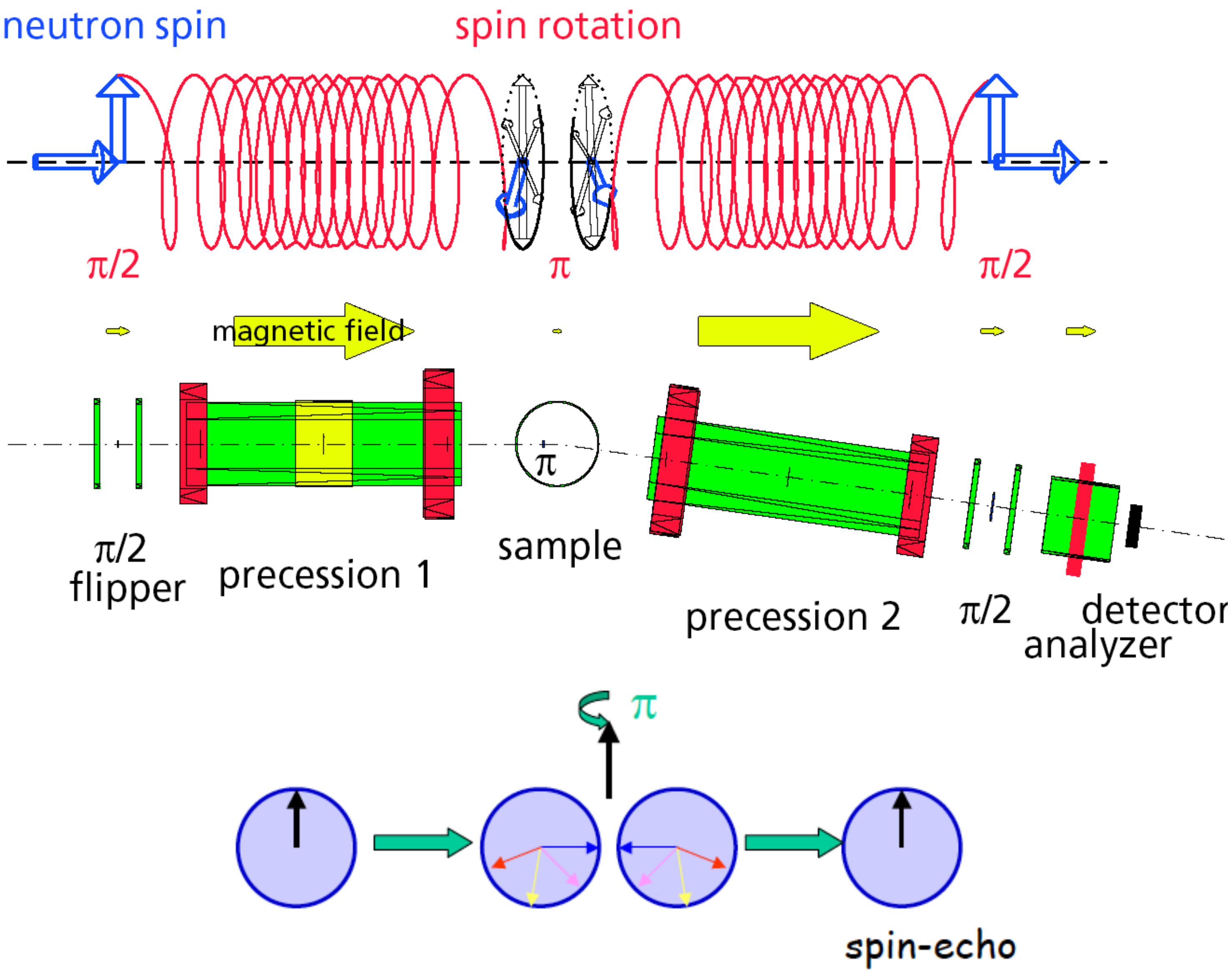
$$\lambda = 8\text{Å}, \ \mathrm{B} = 0.4\mathrm{T}, \ l = 1.2\mathrm{m}$$

$$\rightarrow \varphi/2\pi \cong 3 \times 10^4$$ [Number of Turns]

Notes:

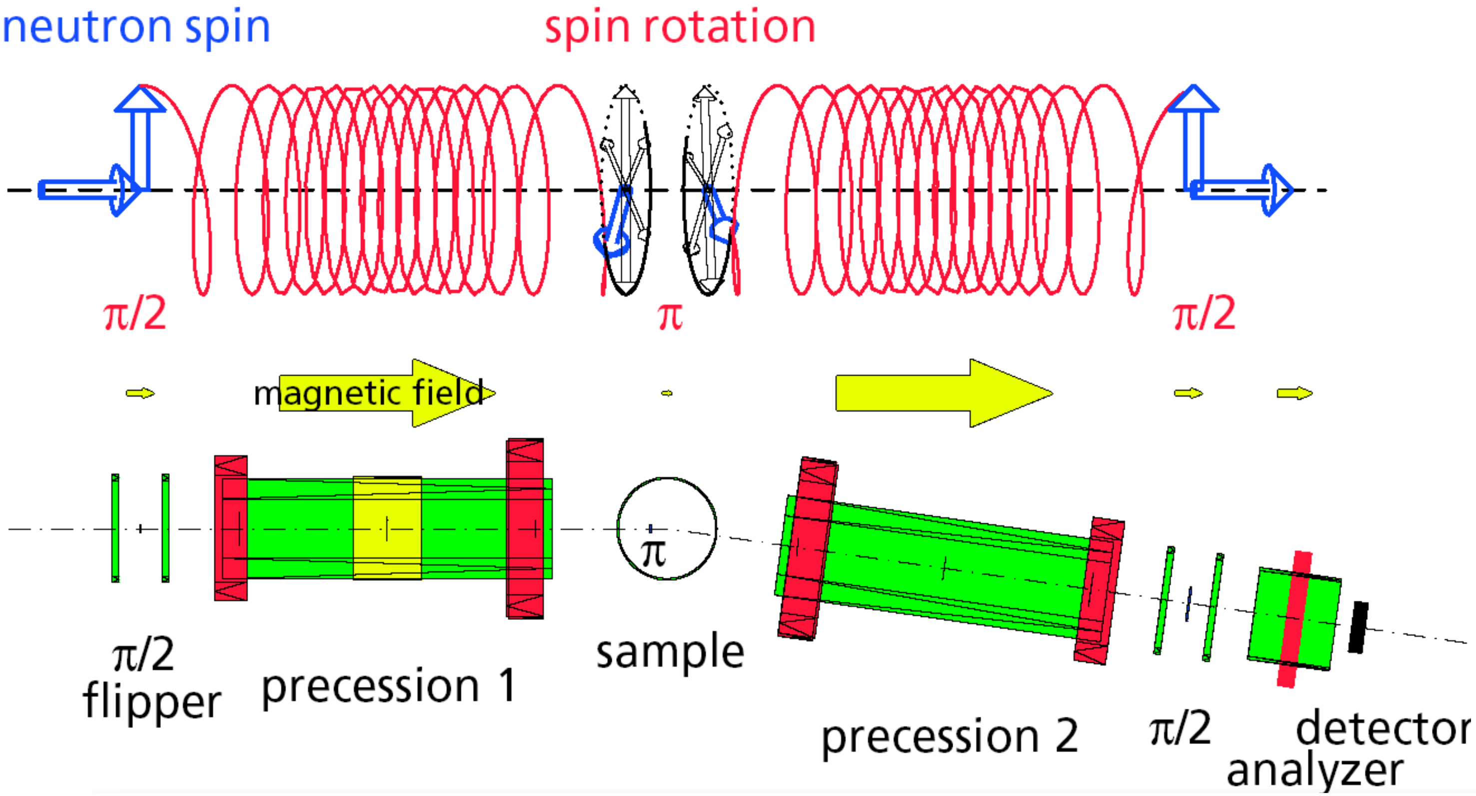
$J \overset{\text{def}}{=} Bl$ or more precisely $J \overset{\text{def}}{=} \int \vec{B} \cdot \vec{dl}$

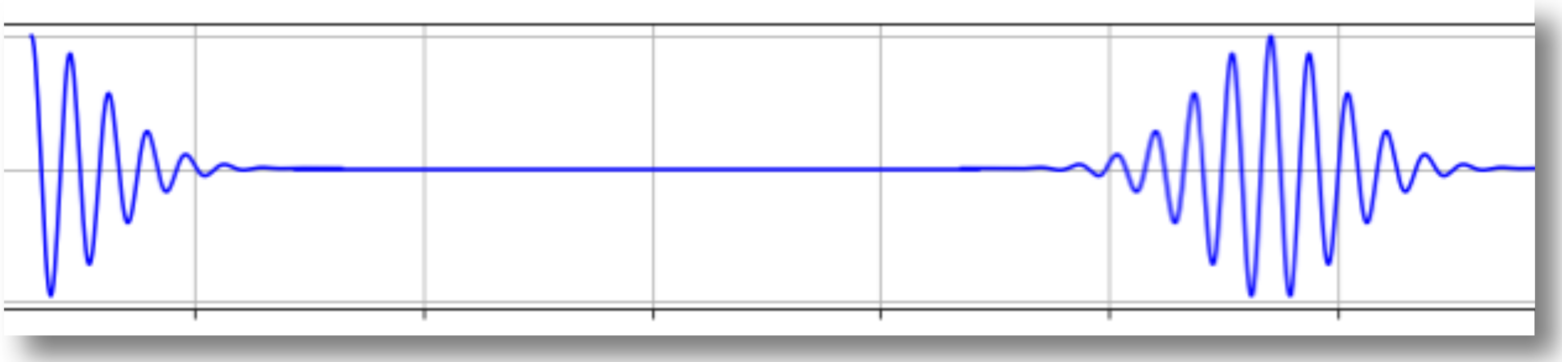
Neutron Spin Echo (I)



Courtesy: M. Monkenbusch, Forschungszentrum Jülich

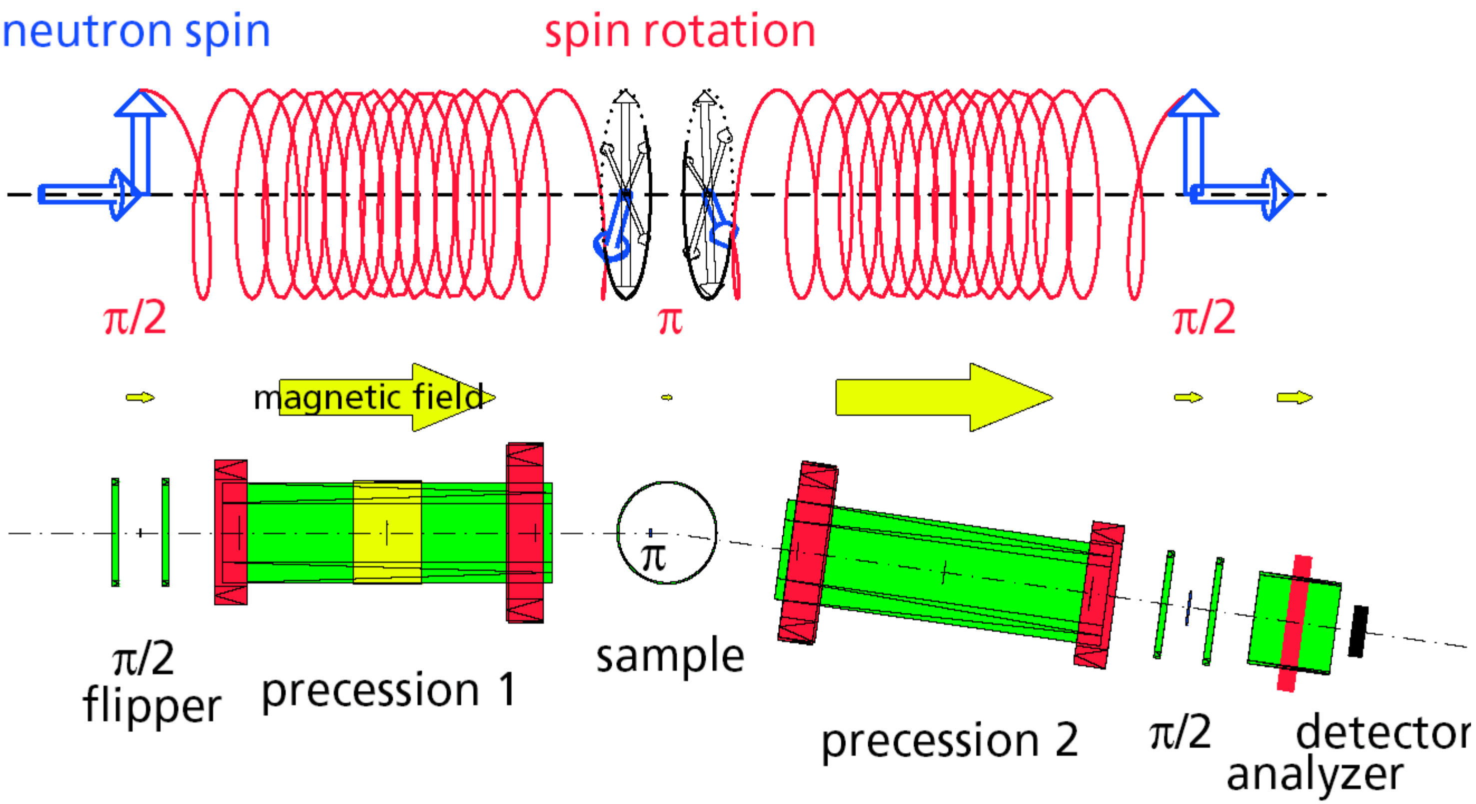
Neutron Spin Echo (II)

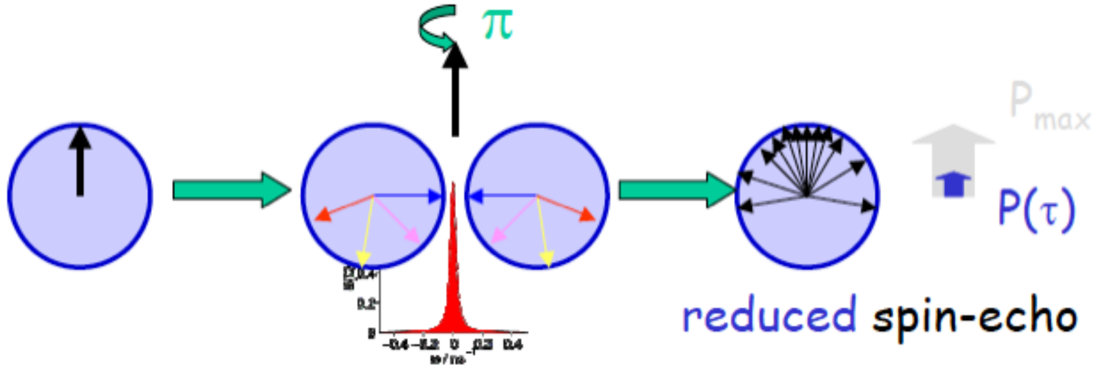




Courtesy: M. Monkenbusch, Forschungszentrum Jülich

Neutron Spin Echo (Inelastic)





Courtesy: M. Monkenbusch, Forschungszentrum Jülich

Neutron Spin Echo

Energy change (quasi-elastic scattering)

$$\Delta E = \hbar\omega = \frac{1}{2}mv_1^2 - \frac{1}{2}mv_2^2 \cong m\, v\, dv$$

Accumulated phase:

$$\phi_2 - \phi_1 = \gamma\left(\frac{B_2 l_2}{v_2} - \frac{B_1 l_1}{v_1}\right) = \gamma B l\left(\frac{1}{v_2} - \frac{1}{v_1}\right) \qquad \Leftarrow \quad B_1 l_1 = B_2 l_2$$

$$\Delta\phi \cong \gamma B l \frac{\hbar\omega}{mv^3} = \frac{\gamma}{2\pi}\left(\frac{m}{h}\right)^2 J\, \lambda^3 \omega$$

or

$$\boxed{\Delta\phi = \tau\omega}$$

where we define Fourier time $\tau \overset{\text{def}}{=} \frac{\gamma}{2\pi}\left(\frac{m}{h}\right)^2 J\, \lambda^3$

For quick computation $\tau \cong 0.186\, J\, \lambda^3$
where $[\tau] = \mathrm{ns}, [J] = \mathrm{Tm}$ and $[\lambda] = \text{Å}$
Example: $\lambda = 8\text{Å},\ J = 0.56\ \mathrm{Tm} \rightarrow \tau \cong 50\mathrm{ns} \rightarrow \Delta E \cong 0.01\ \mu\mathrm{eV}$

Neutron Spin Echo

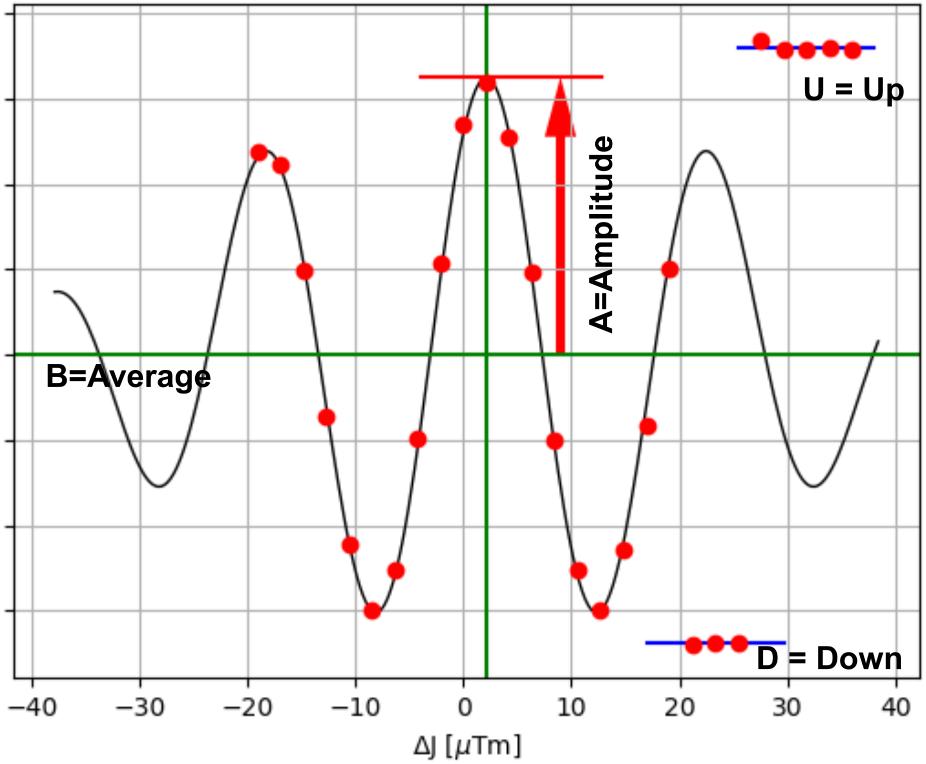
Signal: $I \sim \langle \cos\phi \rangle = \langle \cos \omega\tau \rangle$

$$I \sim \frac{1}{2}\left[S(Q) \pm \int S(Q,\omega)\cos(\omega\tau)\,d\omega\right]$$

Fourier transform
(Real part)

$$B \sim S(Q)$$
$$A \sim I(Q,\tau) = \mathcal{F}[S(Q,\omega)]$$

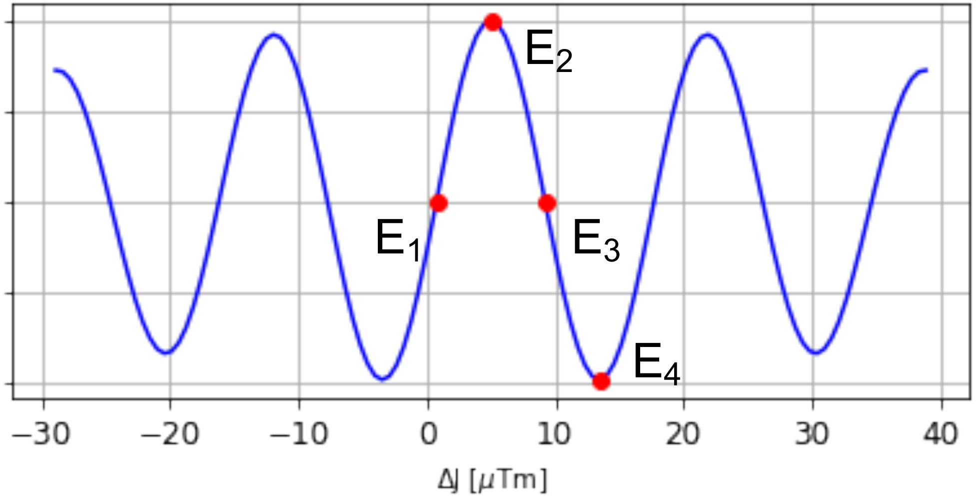
$$\boxed{\frac{I(Q,\tau)}{I(Q,0)} = \frac{2\,A}{U-D}}$$



F. Mezei, Z. Physik (1972) 255: 146.
F. Mezei, C. Pappas, T. Gutberlet (Eds.): Neutron Spin-Echo Spectroscopy, Lecture Notes in Physics 601, Springer, Heidelberg, 2003.

Standard 4-point echo evaluation

$\Delta\lambda/\lambda_0$ ~15-20%



90-degree steps

$$E_1 = B + A\sin(\varphi_0)$$
$$E_2 = B + A\cos(\varphi_0)$$
$$E_3 = B - A\sin(\varphi_0)$$
$$E_4 = B - A\cos(\varphi_0)$$

$$\Rightarrow$$

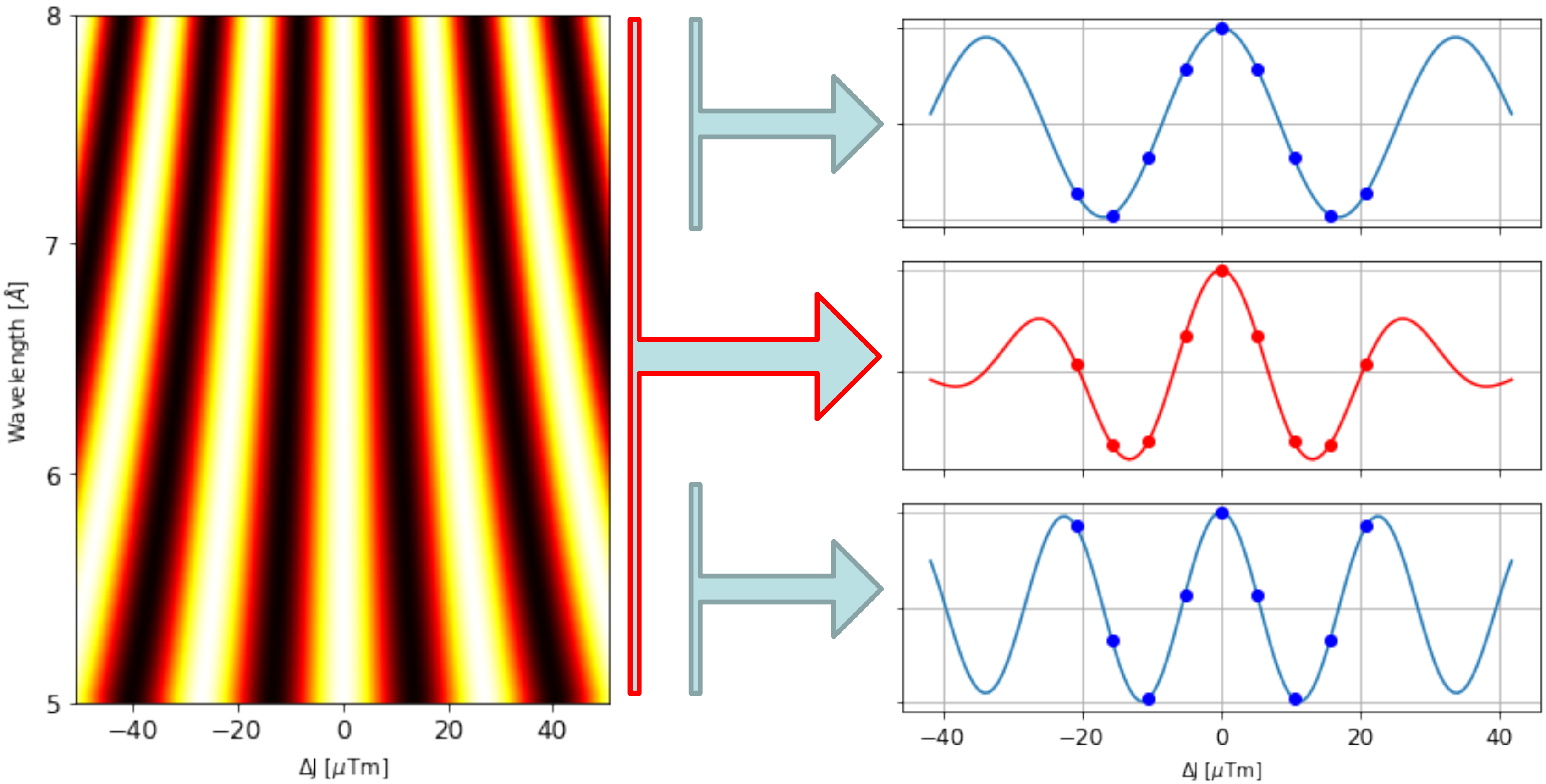
$$B = \frac{1}{4}\sum_{i=1..4} E_i$$

$$A = \sqrt{\frac{1}{2}\sum_{i=1..4}(E_i - B)^2}$$

Time-of-Flight and NSE

$\Delta\lambda/\lambda_0$ up to ~50%

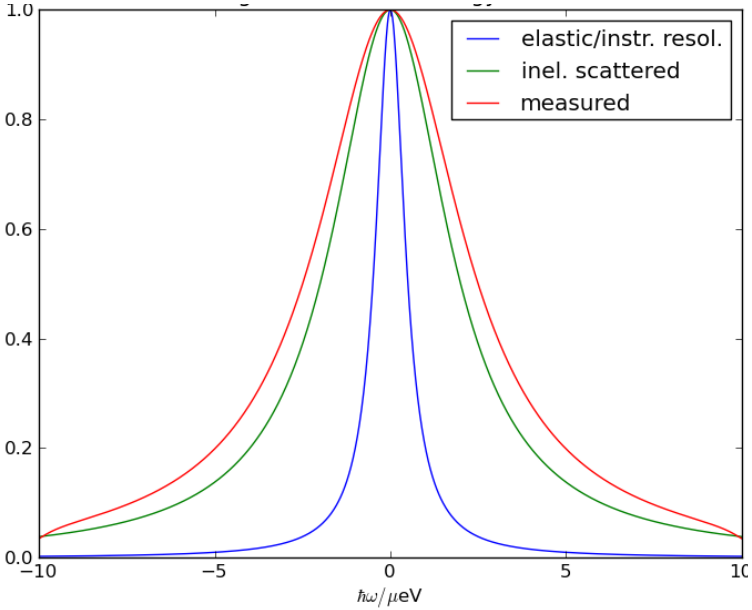
Echo Signals

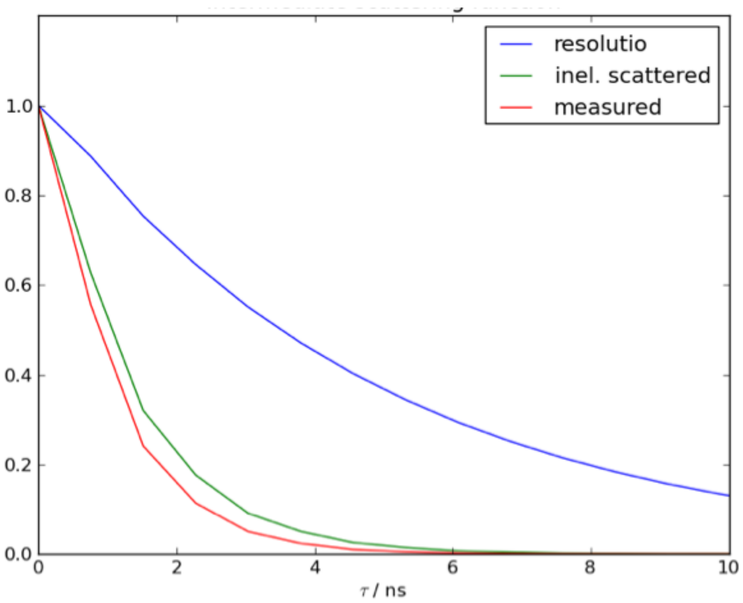


1) F. Mezei, Nucl. Inst. Methods **164**, 153-156 (1979)
2) B. Farago, Time-of-Flight Neutron Spin Echo: Present Status in F.Mezei, C.Pappas, T.Gutberlet Neutron Spin Echo Spectroscopy, Springer (2003)

Energy and Time Domain (Backscattering ↔ NSE)

Dynamic Structure Factor
S(Q,ω) ⟵ *F.T.* ⟶ Intermediate Scattering Function
I(Q,τ)





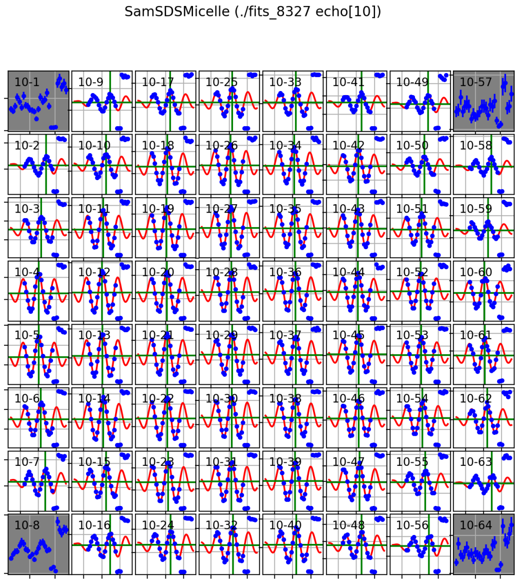
$$I_D(Q,\omega) = S(Q,\omega) * R(Q,\omega)$$

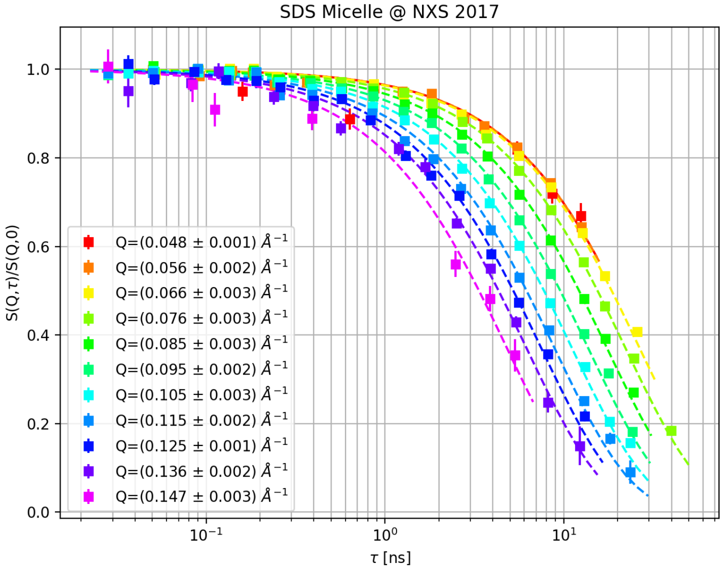
$$I_D(Q,\tau) = I(Q,\tau)\ R(Q,\tau)$$

To get S(Q,ω) we have to
<u>de-convolute</u> instrument resolution

To get I(Q,τ) we just
<u>divide out</u> instrument resolution

Data Reduction

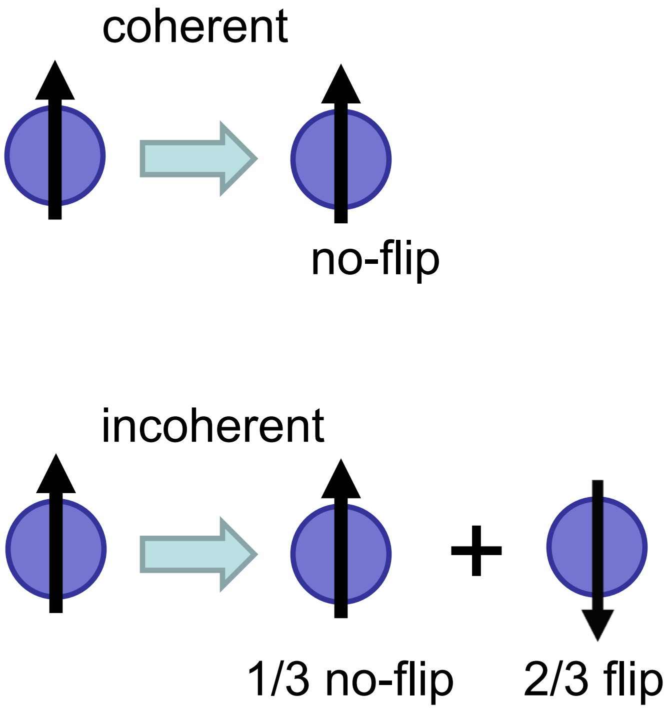




1. Resolution
    - symmetry phase (echo)
    - get $R(Q,\tau;\text{pixel})$
2. Sample: $I_{raw}(Q,\tau;\text{pixel})$
3. Background:
    - $I_{bgr}(Q,\tau;\text{pixel})$
    - correction $\rightarrow I_{sig}(Q,\tau;\text{pixel})$
4. Compute $I(Q,\tau;\text{pixel}) = \frac{\mathrm{I_{sig}}(Q,\tau)}{R(Q,\tau)}$
5. Group results

Complications

Coherent/Incoherent Scattering

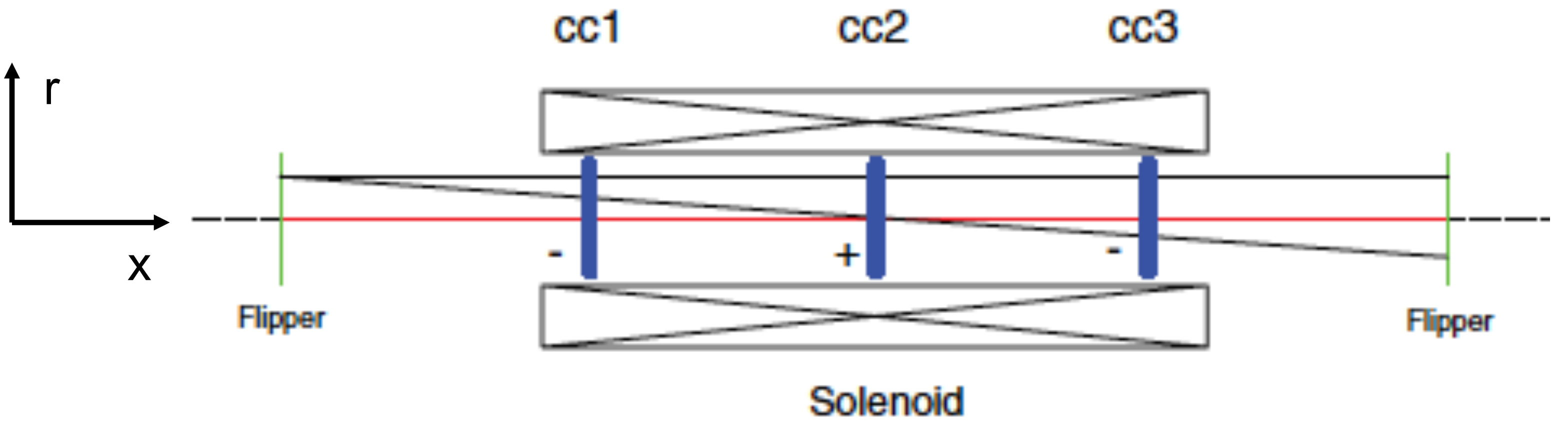


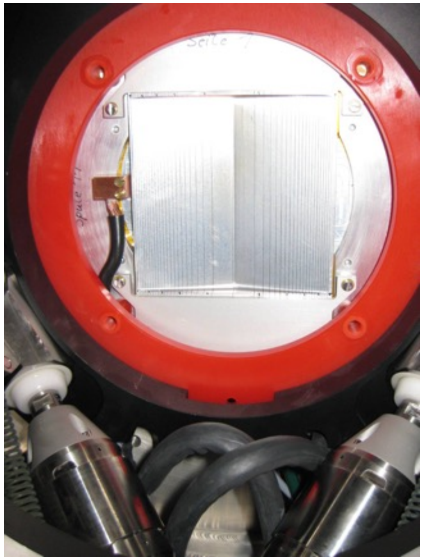
$$I_{\mathrm{up}} = I_{\mathrm{coh}} + \frac{1}{3} I_{\mathrm{inc}}$$

$$I_{\mathrm{down}} = \frac{2}{3} I_{\mathrm{inc}}$$

$$I_{\mathrm{amp}} = I_{\mathrm{coh}} f_{\mathrm{coh}}(\tau) - \frac{1}{3} I_{\mathrm{inc}} f_{\mathrm{inc}}(\tau)$$

Correction Coils



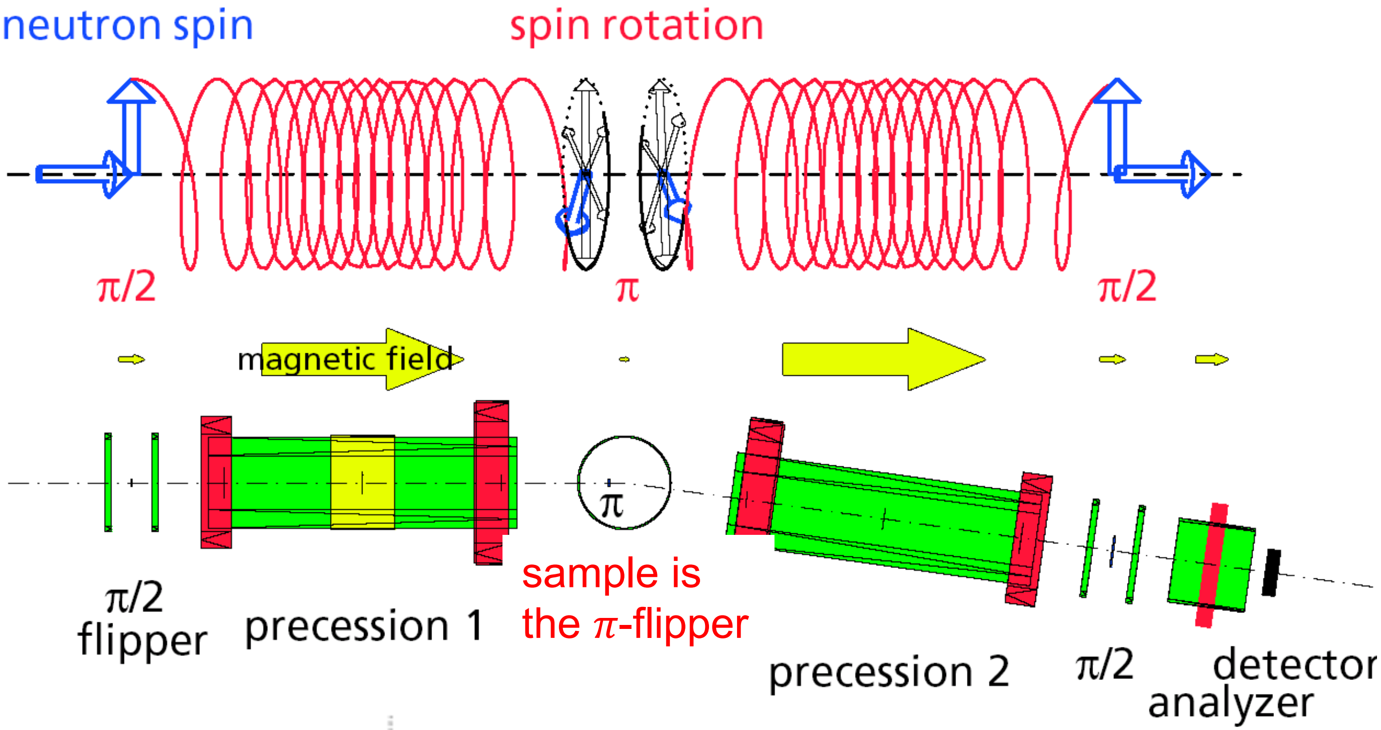


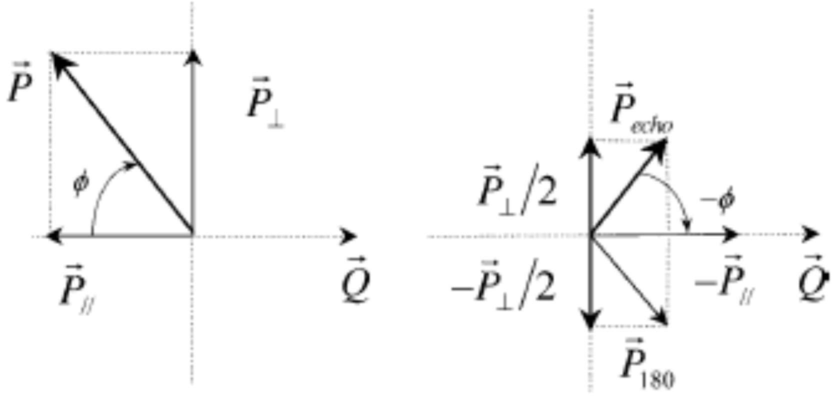
- Problem:
  - Field integral $J = Bl$ the same for all trajectories
  - Solenoid field $B(r) \sim r^2$
- Solution:
  - correction coils with current distribution that varies as r^2

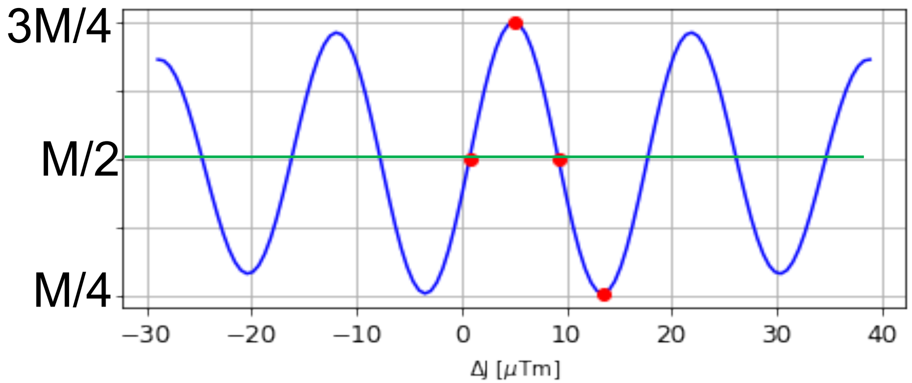
S. Pasini, M. Monkenbusch, Meas. Sci. Technol. 26 (2015) 035501

Theme Variations

Paramagnetic NSE



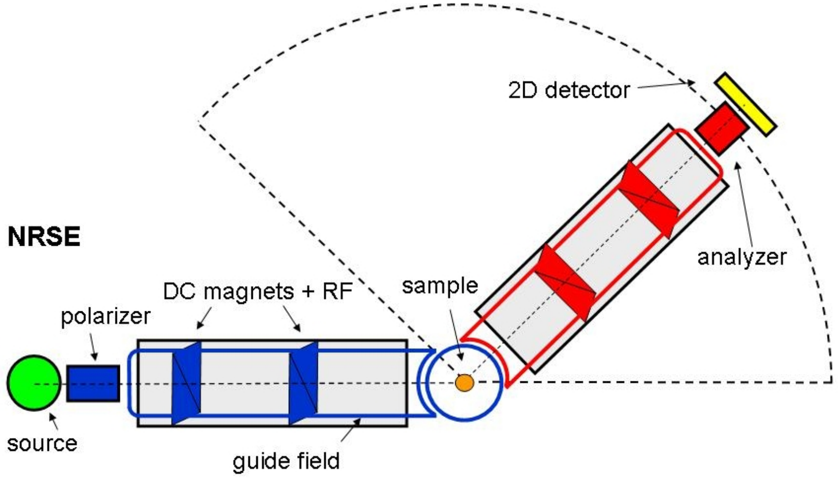




C.Pappas, G. Ehlers, F. Mezei, Neutron Spin Echo and Magnetism in Neutron Scattering from Magnetic Materials, ed. T. Chatterji (2006)

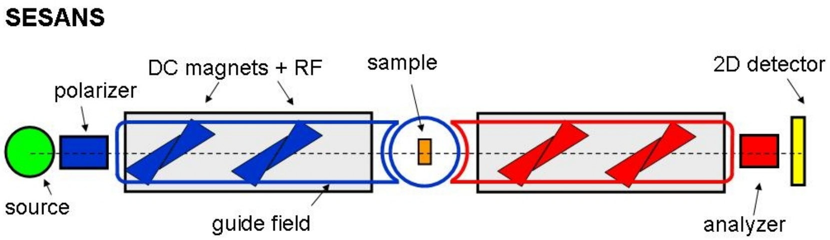
“Non-standard” NSE Instruments

NSRE



RF field instead of solenoid

SESANS



Spin Echo encoded SANS

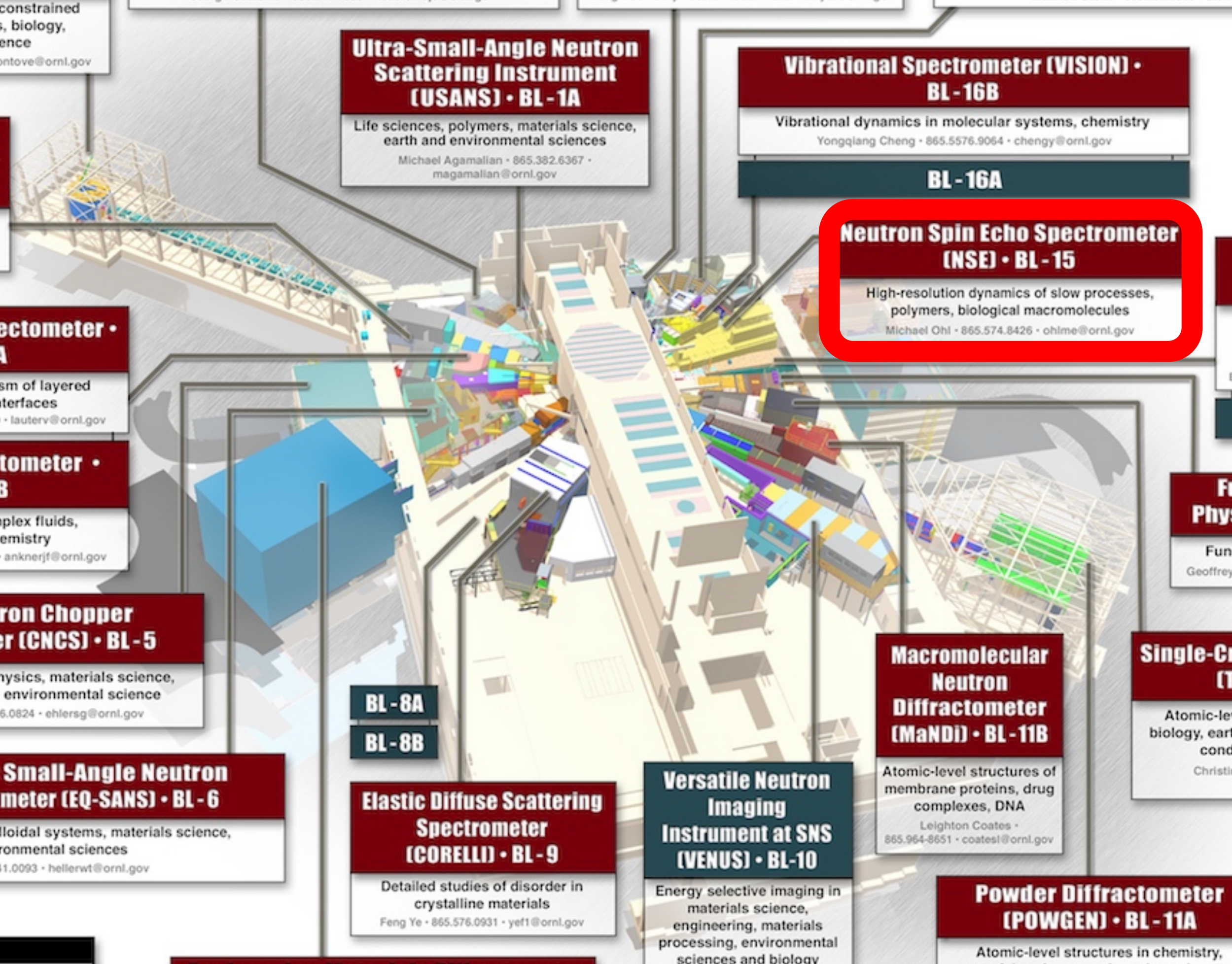
SNS-NSE: NSE Spectrometer at SNS

OAK RIDGE
National Laboratory

SPALLATION
NEUTRON
SOURCE

World’s most intense pulsed, accelerator-based neutron source

NEUTRONS.ORNL.GOV



Backscattering Spectrometer (BASIS) • BL - 2

Dynamics of macromolecules, constrained molecular systems, polymers, biology, chemistry, materials science

Eugene Mamontov • 865.574-5109 • mamontove@ornl.gov

Nanoscale-Ordered Materials Diffractometer (NOMAD) • BL - 1B

Liquids, solutions, glasses, polymers, nanocrystalline and partially ordered complex materials

Joerg Neuefeind • 865.241.1635 • neuefeindjc@ornl.gov

Wide Angular-Range Chopper Spectrometer (ARCS) • BL - 18

Atomic-level dynamics in materials science, chemistry, condensed matter sciences

Doug Abernathy • 865.617.5624 • abernathydl@ornl.gov

Fine-Resolution Fermi Chopper Spectrometer (SEQUOIA) • BL - 17

Dynamics of complex fluids, quantum fluids, magnetism, condensed matter, materials science

Matthew Stone • 865.202.6898 • stonemb@ornl.gov

Ultra-Small-Angle Neutron Scattering Instrument (USANS) • BL - 1A

Life sciences, polymers, materials science, earth and environmental sciences

Michael Agamalian • 865.382.6367 • magamalian@ornl.gov

Vibrational Spectrometer (VISION) • BL - 16B

Vibrational dynamics in molecular systems, chemistry

Yongqiang Cheng • 865.5576.9064 • chengy@ornl.gov

BL - 16A

Spallation Neutrons and Pressure Diffractometer (SNAP) • BL - 3

Materials science, geology, earth and environmental sciences

Chris Tulk • 865.574.5764 • tulkca@ornl.gov

Neutron Spin Echo Spectrometer (NSE) • BL - 15

High-resolution dynamics of slow processes, polymers, biological macromolecules

Michael Ohl • 865.574.8426 • ohlme@ornl.gov

Hybrid Spectrometer (HYSPEC) • BL - 14B

Atomic-level dynamics in single crystals, magnetism, condensed matter sciences

Barry Winn • 865.805.6819 • winnbl@ornl.gov

BL - 14A

Magnetism Reflectometer • BL - 4A

Chemistry, magnetism of layered systems and interfaces

Valeria Lauter • 865.387.5389 • lauterv@ornl.gov

Liquids Reflectometer • BL - 4B

Interfaces in complex fluids, polymers, chemistry

John Ankner • 865.377.4523 • anknerjf@ornl.gov

Fundamental Neutron Physics Beam Line • BL - 13

Fundamental properties of neutrons

Geoffrey Greene • 865.576.0023 • greenegl@ornl.gov

Cold Neutron Chopper Spectrometer (CNCS) • BL - 5

Condensed matter physics, materials science, chemistry, biology, environmental science

Georg Ehlers • 865.206.0824 • ehlersg@ornl.gov

BL - 8A

BL - 8B

Macromolecular Neutron Diffractometer (MaNDi) • BL - 11B

Atomic-level structures of membrane proteins, drug complexes, DNA

Leighton Coates • 865.964-8651 • coatesl@ornl.gov

Single-Crystal Diffractometer (TOPAZ) • BL - 12

Atomic-level structures in chemistry, biology, earth science, materials science, condensed matter physics

Christina Hoffmann • 865.576.5127 • choffmann@ornl.gov

Extended Q-Range Small-Angle Neutron Scattering Diffractometer (EQ-SANS) • BL - 6

Life science, polymer and colloidal systems, materials science, earth and environmental sciences

William Heller • 865.241.0093 • hellerwt@ornl.gov

Elastic Diffuse Scattering Spectrometer (CORELLI) • BL - 9

Detailed studies of disorder in crystalline materials

Feng Ye • 865.576.0931 • yef1@ornl.gov

Versatile Neutron Imaging Instrument at SNS (VENUS) • BL-10

Energy selective imaging in materials science, engineering, materials processing, environmental sciences and biology

Hassina Bilheux • 865.384.9630 • bilheuxhn@ornl.gov

Powder Diffractometer (POWGEN) • BL - 11A

Atomic-level structures in chemistry, materials science, and condensed matter physics including magnetic spin structures

Ashfia Huq • 630.986.7321 • huqa@ornl.gov

Engineering Materials Diffractometer (VULCAN) • BL - 7

Mechanical behaviors, materials science, materials processing

Ke An • 865.919.5226 • kean@ornl.gov

*Scheduled commissioning date

LEGEND

🟥 Operating instrument in user program

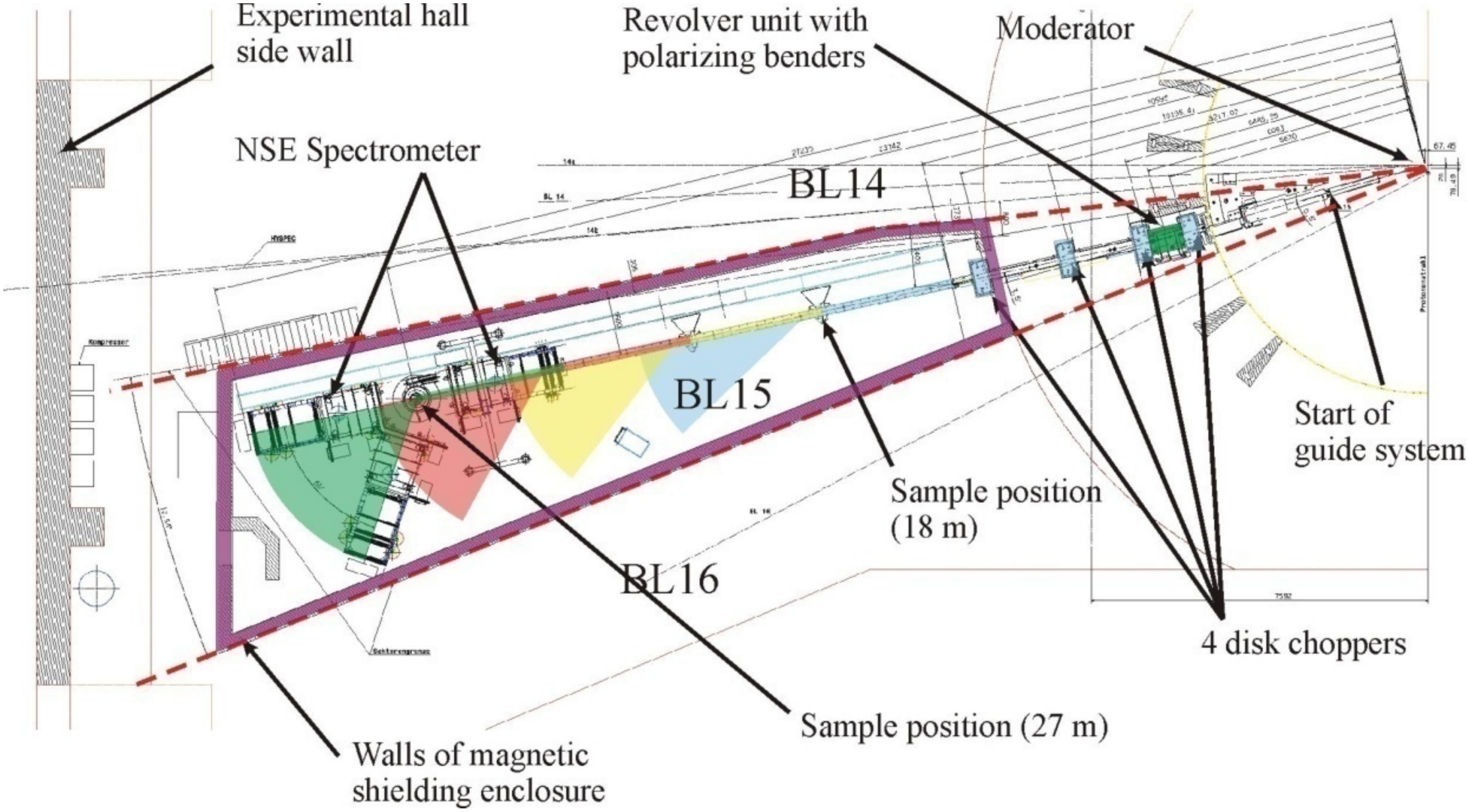
🟪 In design or construction

🟦 Under consideration

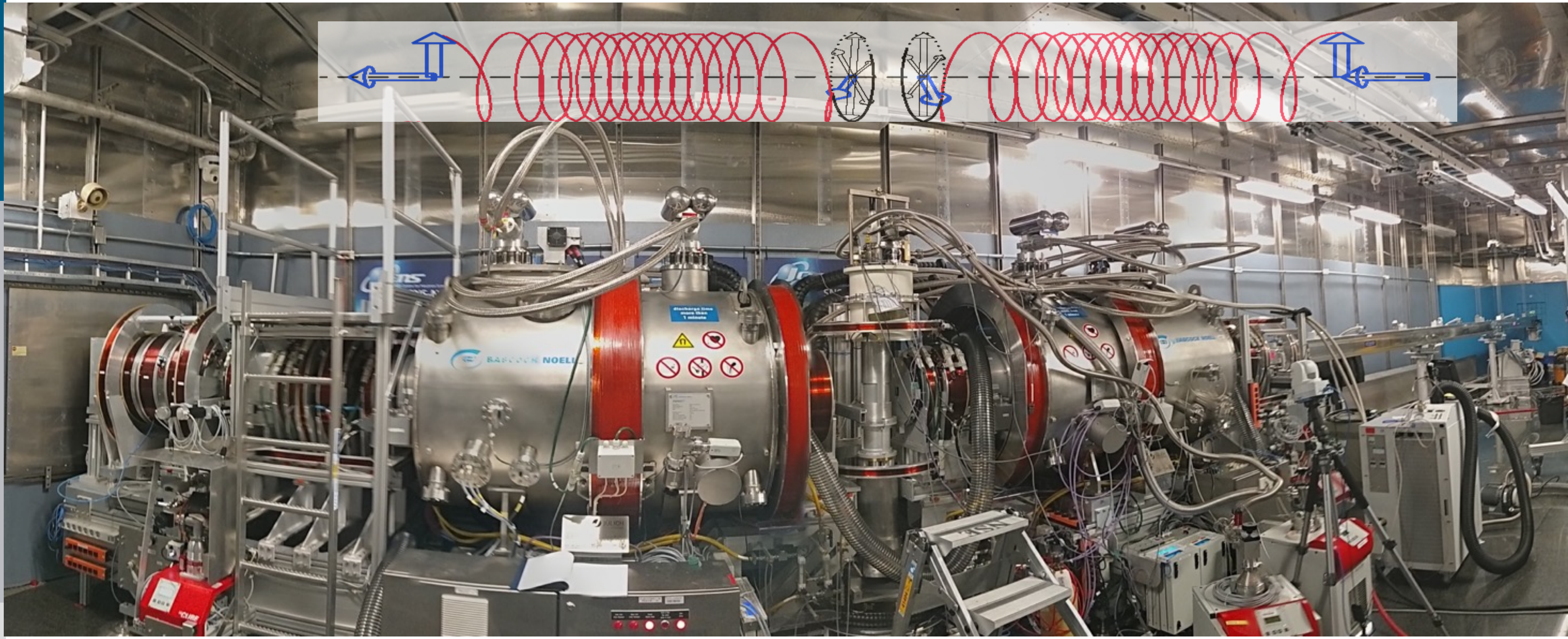
15-G00337A/gim

The Spallation Neutron Source is a facility of Oak Ridge National Laboratory, managed by UT-Battelle for the US Department of Energy.

Instrument Location & Layout



SNS-NSE: IN11-Type NSE

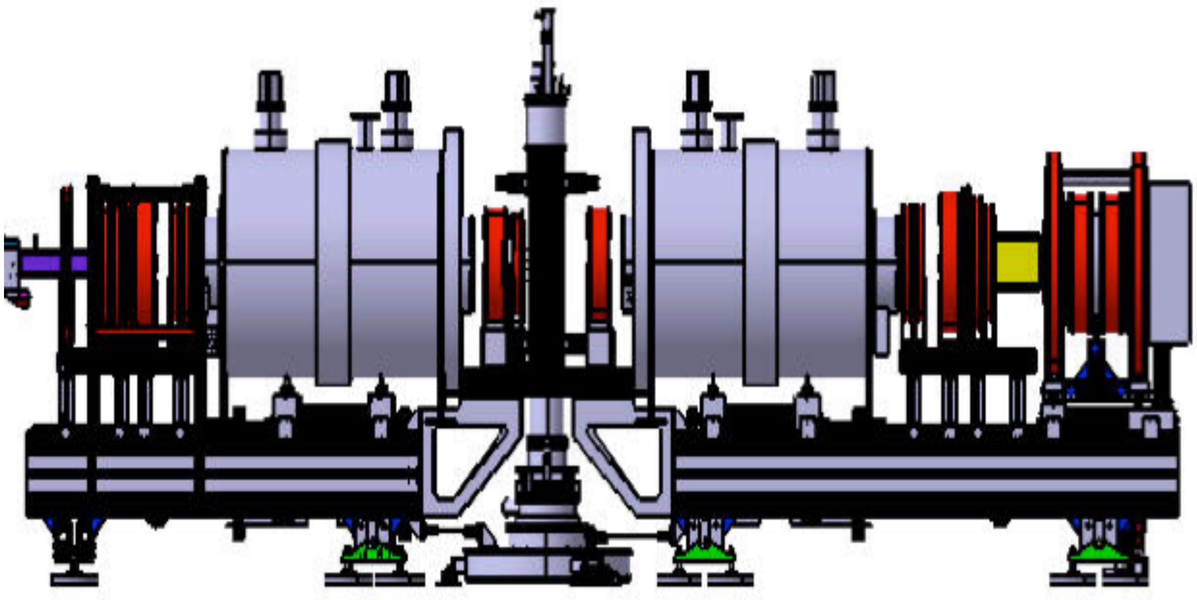


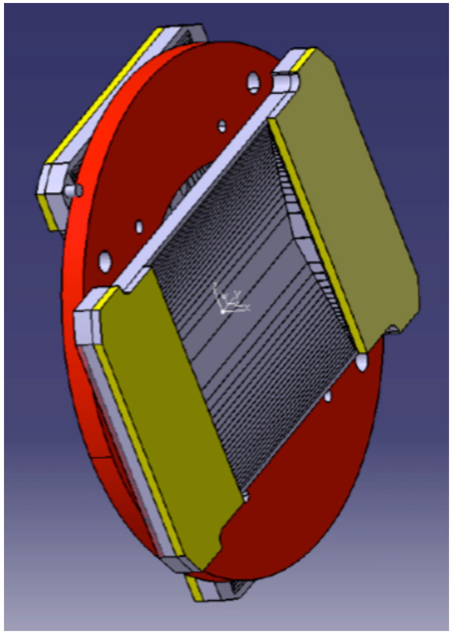
Typical neutron wavelengths:
$\lambda = 2 - 14$ Å, $\Delta\lambda = 2.5, 3.6$ Å

$J_{max} \sim 0.56$ Tm
$2\theta = 3.5^\circ - 79.5^\circ$

SNS-NSE Coils

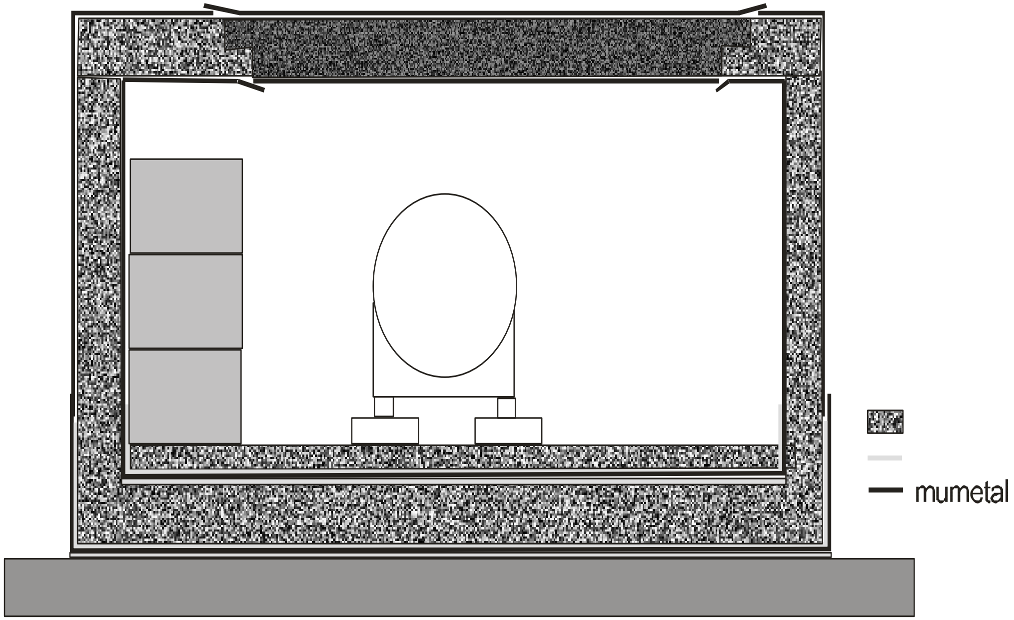
SNS-NSE: the first NSE with superconducting main coils

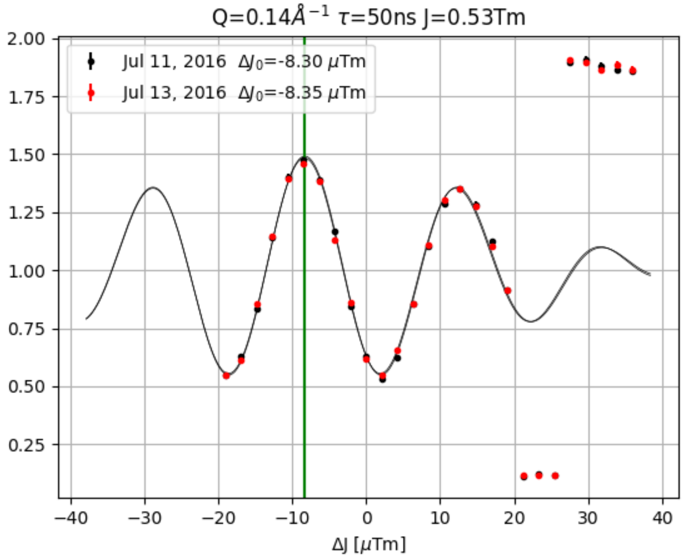




“Pythagorean” correction coils: $r^2 = y^2 + z^2$

Magnetic Shielding





$\Delta\phi \rightarrow 1$ deg

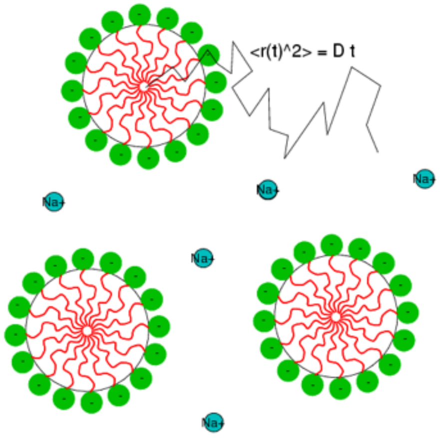
Examples

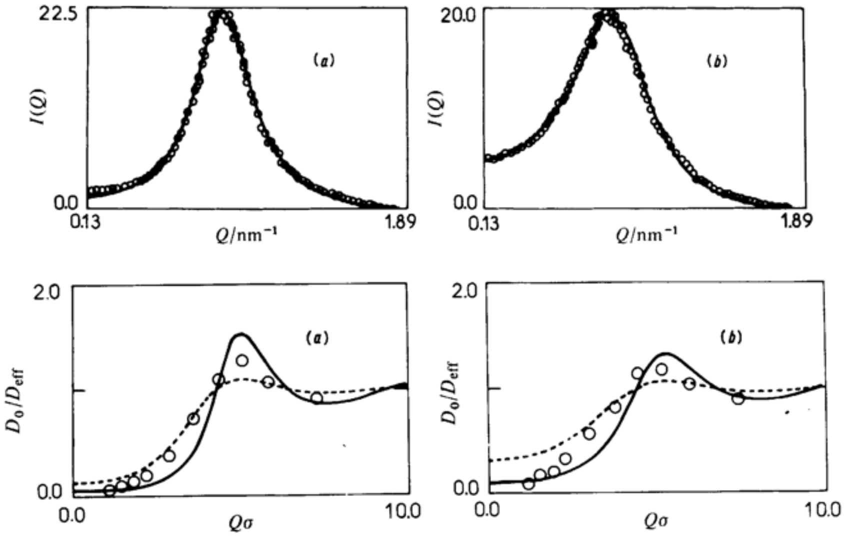
Two ”historical” examples:

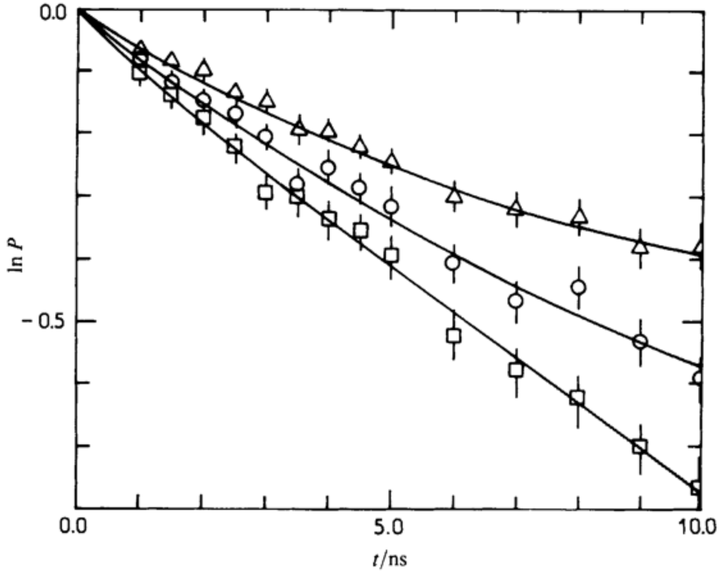
1. SDS Micelles (1981)
2. Reptation Model (2002)

Several recent results from SNS-NSE

SDS Micelles







$$\frac{I(Q,t)}{I(Q,0)} = e^{-D_{\mathrm{eff}}(Q)Q^2 t}$$

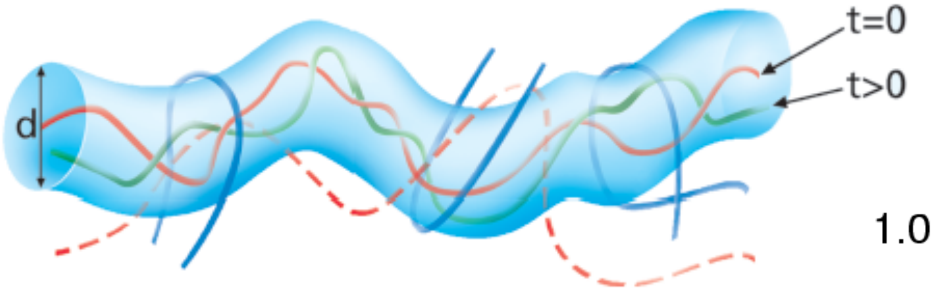
$$D_{\mathrm{eff}} = D_0 H(Q)/S(Q)$$

$$D_0 = \frac{kT}{6\pi\eta R}$$

J. Hayter, J. Penfold, J. Chem. Soc., Faraday Trans. 1, 1981,77, 1851-1863

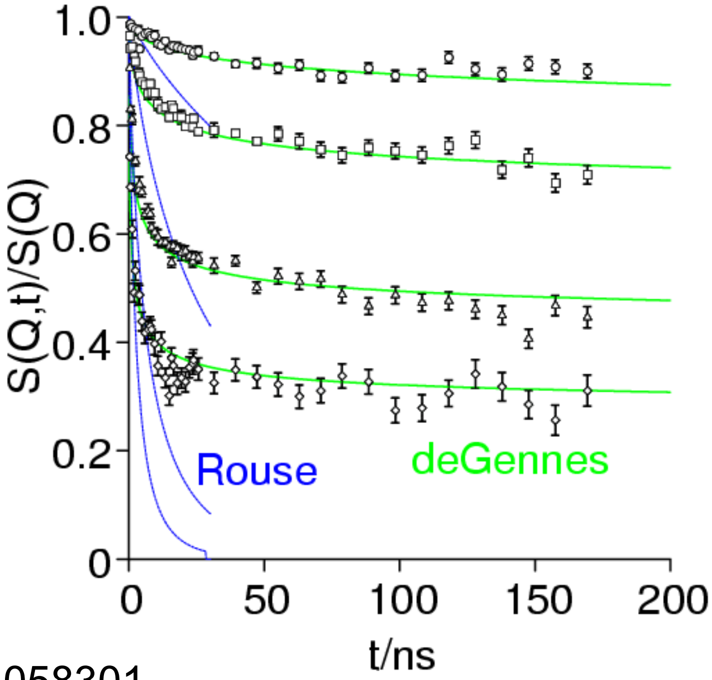
Reptation Model

(de Gennes/Edwards)



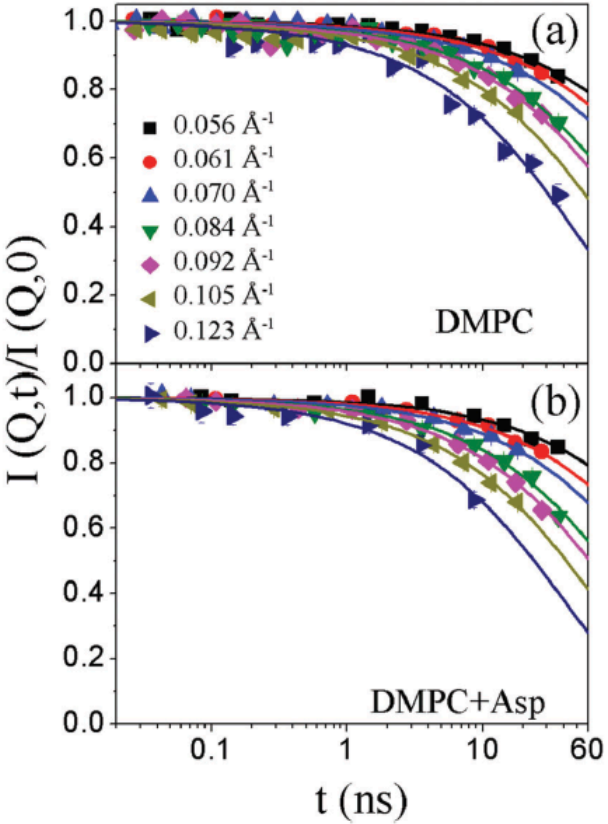
Coherent scattering, labeled chain

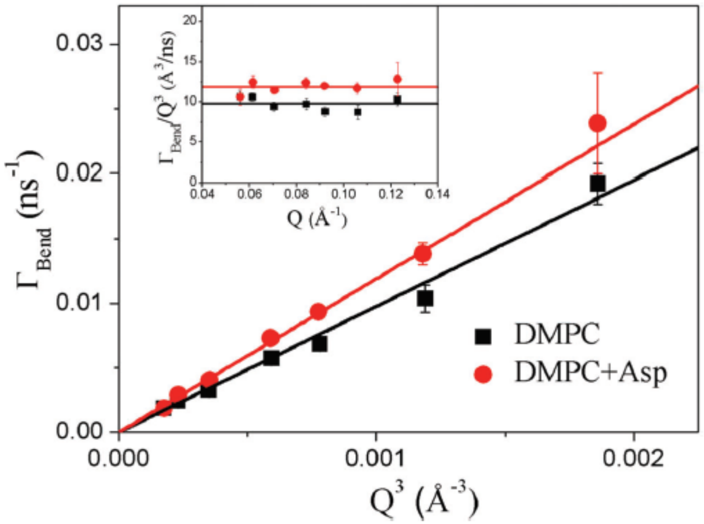
Melt of long chain linear PE



A. Wischnewski *et al.*, Phys. Rev. Lett. **88** (2002), 058301

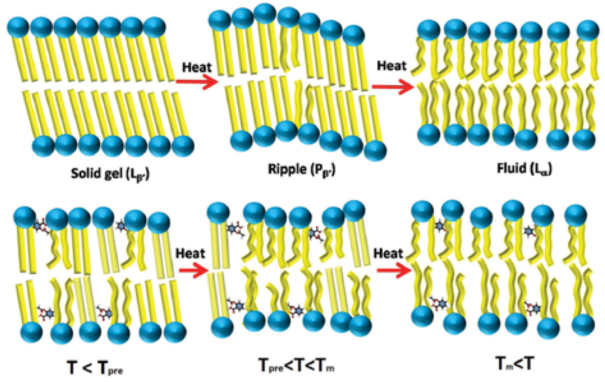
Aspirin modulates the dynamical behavior of membranes





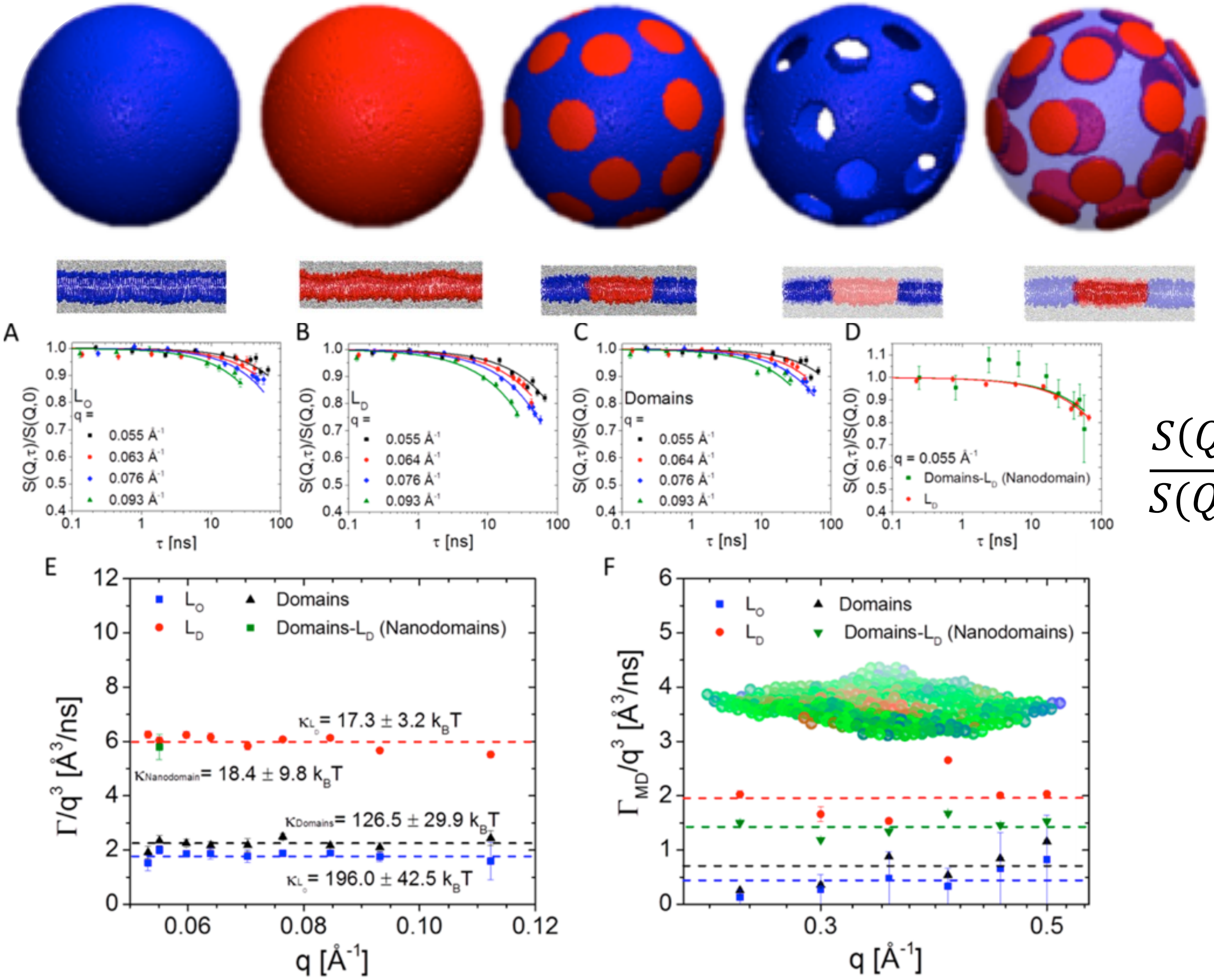
$$\frac{I(Q,t)}{I(Q,0)} = \exp(-(\Gamma_{\text{Bend}} t)^{2/3})$$

$$\Gamma_{\text{Bend}} = 0.025\,\gamma_k \left(\frac{k_B T}{\kappa}\right)^{1/2} \left(\frac{k_B T}{\eta}\right) Q^3$$



V. K. Sharma et al., Phys. Chem. Chem. Phys. 2017

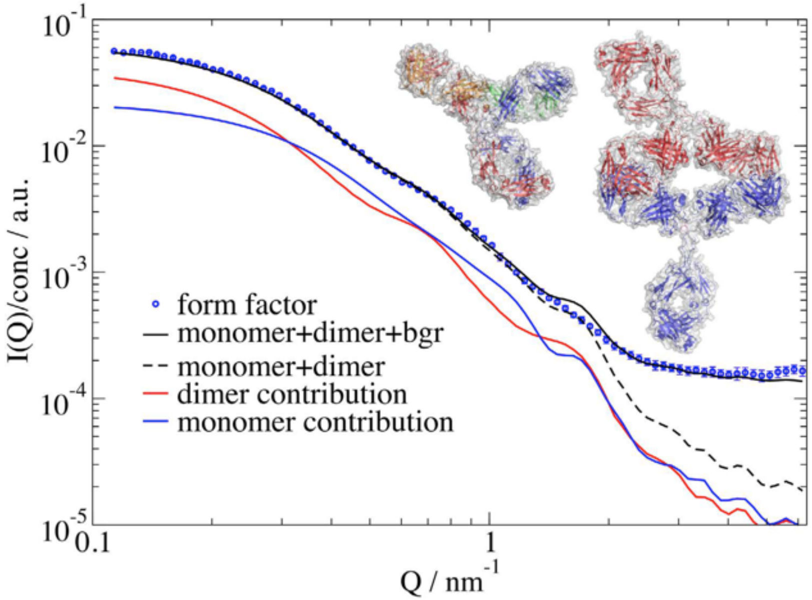
Mechanical Properties of Nanoscopic Lipid Domains



$$\frac{S(Q,t)}{S(Q,0)} = \exp(-(\Gamma(Q)t)^{2/3})$$

J. Nickels et al., J. Am. Chem. Soc., **2015**, *137* (50)

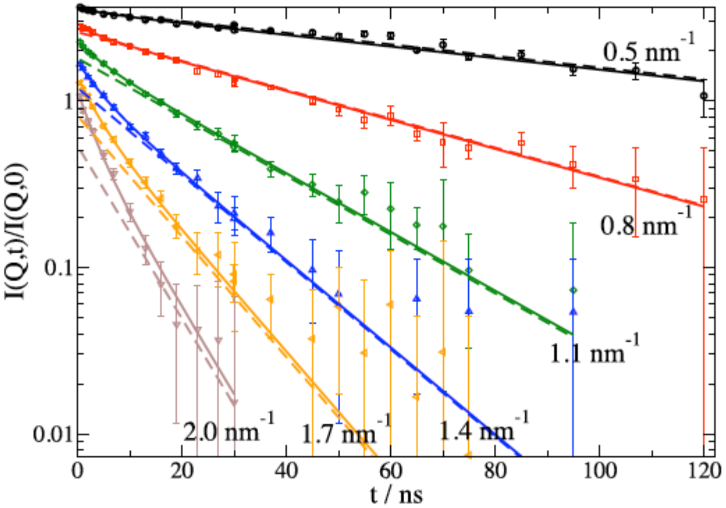
Fast antibody domain motion

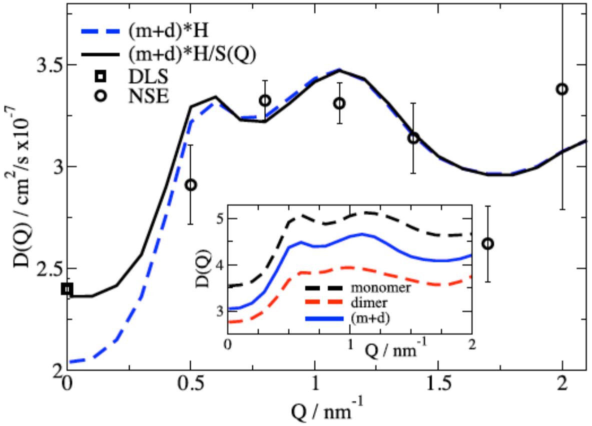


$$F(Q,t) = F_{trans}(Q,t) \cdot F_{rot}(Q,t) \cdot F_{int}(Q,t)$$

$$F_{trans}(Q,t) = \exp(-Q^2 D_T t)$$

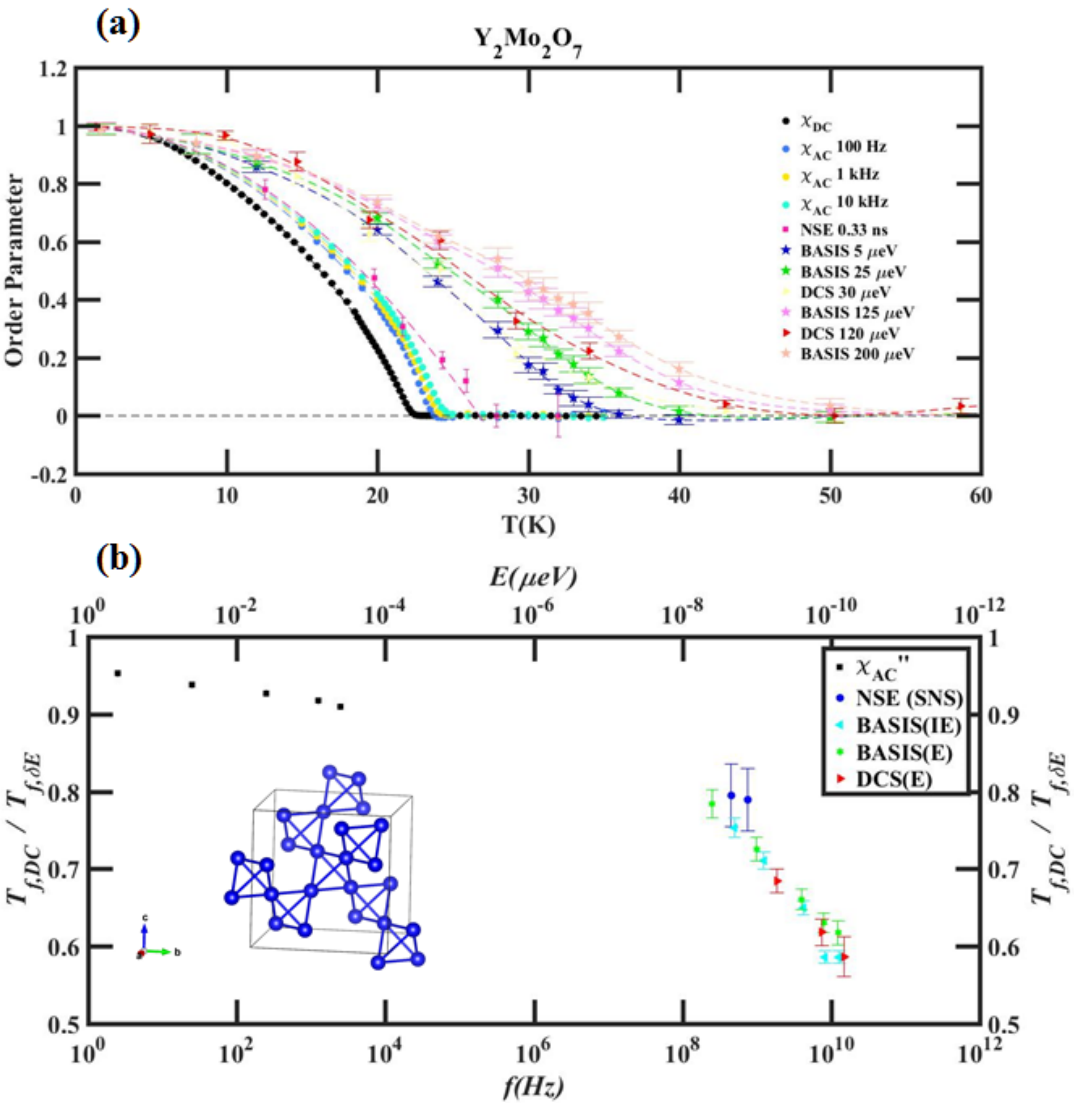
$$F_{int}(Q,t) = \left\langle \sum_{\alpha,\beta} b_\alpha b_\beta \exp(iQR_\beta^{eq}) \exp(-iQR_\alpha^{eq}) \cdot f_{\alpha\beta}(Q,\infty) \cdot f'_{\alpha\beta}(Q,t) \right\rangle$$





L.R. Stingaciu et al. Scientific Reports 2016

Example of Paramagnetic NSE



Courtesy of A. Samarakoon, University of Virginia

Summary

- NSE
  - a high resolution neutron spectroscopy
  - measures the intermediate scattering function S(Q,τ)
  - need large samples, scattering intensity

- SNS-NSE (BL-15)
  - the first NSE at a Spallation Source
  - the first one with superconducting coils
  - the only one with magnetic shielding
  - available in user program – write a proposal!!

Acknowledgements: M. Monkenbusch, D. Richter, L. Stingaciu, M. Cochran, G. Ehlers, W-R Chen, T. Kozielewski, M. Ohl, N. Arend and M.Sharp

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   http://doi.org/10.1016/j.crhy.2007.10.001
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   https://www.ill.eu/fileadmin/users_files/documents/links/documentation/NeutronDataBooklet.pdf
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