Interpolative Decomposition and its Applications in Quantum Chemistry

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June 19-21, 2018, CSCAMM, UMD
Mathematical and Numerical Aspects of Quantum Dynamics

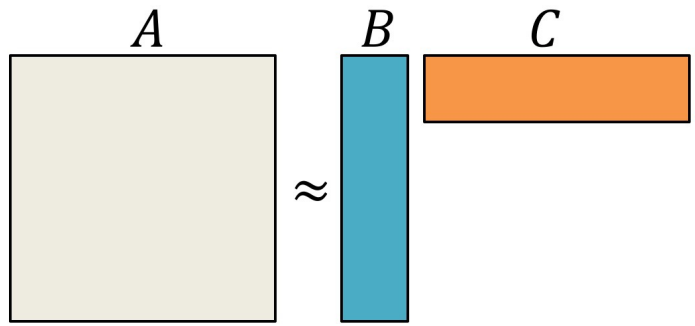
Outline

- What is interpolative decomposition?
  - Some theory
  - How to compute it
- Kohn-Sham density functional theory
- Applications
  - Localization
  - Electron repulsion integral tensor

Low-rank approximation

- Given a matrix $A \in \mathbb{R}^{m \times n}$, a low-rank approximation of A is

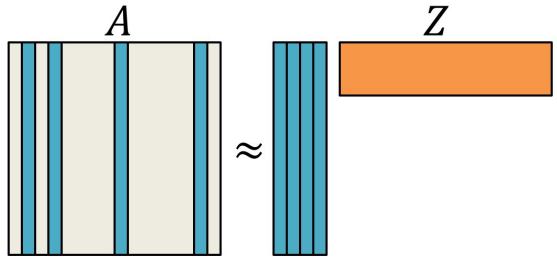
$$A \approx BC, \quad B \in \mathbb{R}^{m \times k}, C \in \mathbb{R}^{k \times n}, k \ll \min(m, n).$$



- Example: singular value decomposition $A \approx (US)V^T$.
- Applications
  - Principal component analysis,
  - Signal processing (compression, denoising, ...)
  - Fast numerical linear algebra
  - Sparse recovery (collaborative filtering)

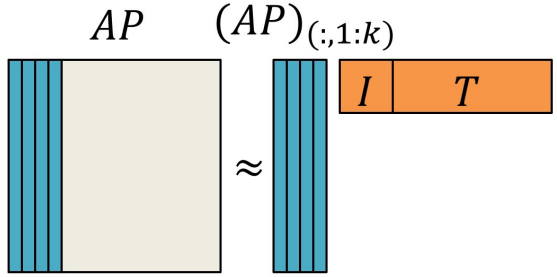
Interpolative decomposition (ID)

- A low-rank approximation that uses A's own columns.



- The picked columns are called the skeletons.
- Let P be the permutation matrix moving the skeletons to the front.

$$AP \approx (AP)_{(:,1:k)} \begin{bmatrix} I & T \end{bmatrix}, \quad T \in \mathbb{R}^{k \times (n-k)}.$$



- Key advantages: keep the columns of A in the approximation
  - Reuse the entries of A (save space),
  - Keep structure of the columns.

Theory

Theorem (Gu+Eisenstat, Tyrtyshnikov)

For fixed k, there exists P and T s.t.

- $\left\| AP - (AP)_{(:,1:k)} \begin{bmatrix} I & T \end{bmatrix} \right\|_2 \leq \sqrt{1 + k(n-k)}\sigma_{k+1}(A)$,
- $|T_{ij}| \leq 1$.

Proof.

- Find k columns of A that span the maximal volume. This implies that $|T_{ij}| \leq 1$.
- Build a QR decomposition based on these columns to derive the error bound.

□

- $\sigma_{k+1}(A)$ is the best approximation result from SVD. ID has an extra $\sqrt{nk}$ factor (in the worst case).
- Complexity: combinatorial search, exponential cost.

Theory

Theorem (Gu+Eisenstat)

For fixed k and fixed $f > 1$, there exists P and T s.t.

- $\left\| AP - (AP)_{(:,1:k)} \begin{bmatrix} I & T \end{bmatrix} \right\|_2 \leq \sqrt{1 + f^2 k(n-k)}\sigma_{k+1}(A)$,
- $|T_{ij}| \leq f$,

and it can be found in $O((m + n \log_f n)n^2)$ steps.

Proof.

- Iteratively improve the column selection by finding the largest entry T_{ij} with $|T_{ij}| > f$.
- Number of iterations bounded by $O(\log_f n^{k/2}) = O(k \log_f n)$.
- Efficient routines for updating the factorization once a new column is picked.

□

- Approx. error has an extra $\sqrt{f^2 nk}$ factor compared to SVD.
- Complexity: for $f = O(n^\alpha)$, the cost is cubic $O(mn \min(m, n))$.

In practice

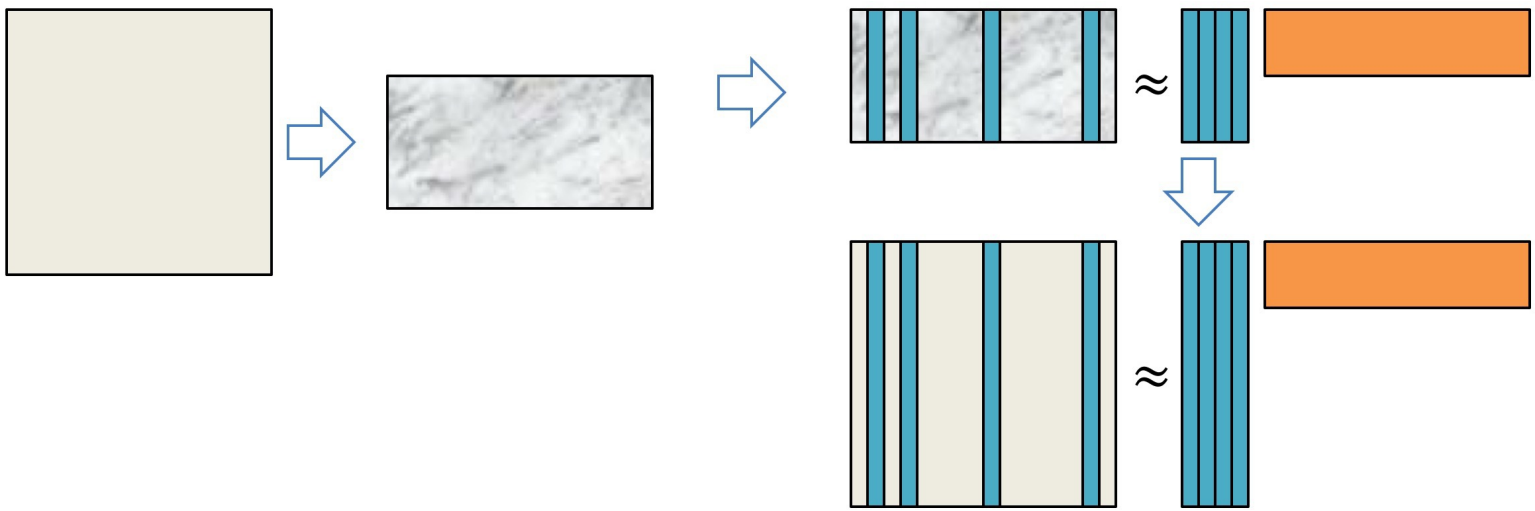
- QR with column pivoting (QRCP).
  - A greedy heuristic for maximizing the volume of the picked columns one by one.

$$AP = QR = \begin{bmatrix} Q_1 & Q_2 \end{bmatrix} \begin{bmatrix} R_{11} & R_{12} \\ & R_{22} \end{bmatrix} \approx Q_1 \begin{bmatrix} R_{11} & R_{12} \end{bmatrix}$$
$$\approx (Q_1 R_{11}) \begin{bmatrix} I & R_{11}^{-1} R_{12} \end{bmatrix} := (AP)_{(:,1:k)} \begin{bmatrix} I & T \end{bmatrix}.$$

- Almost the same cost of standard QR: $O(mnk)$.
- No guarantee for a bound on $|T_{ij}|$ but works well in most cases.

In practice

- Randomized approach if $\min(m, n) \gg k$
  - Project the columns (via randomized Fourier transform [Ailon-Chazelle-2009]) to a random $O(k)$ dimensional subspace.
  - Apply QRCP to the projected (fat) matrix.



- Benefits
  - Reuse the entries of A (save space),
  - Inherit the structure of the columns: sparsity, locality, factorized form.

Extension

- Two-sided interpolative decomposition [Cheng et al.-2006]

$$P^{\mathsf{T}} A Q \approx \begin{bmatrix} I \\ T_1^{\mathsf{T}} \end{bmatrix} (P^{\mathsf{T}} A Q)_{(1:k,1:k)} \begin{bmatrix} I & T_2 \end{bmatrix}.$$

  - Apply QRCP to both the rows and the columns.
  - Can be combined with the randomization.

- In what follows, we assume that the columns are already in the correct order for ID (i.e., $P = I$)

$$A \approx A_{(:,1:k)} \begin{bmatrix} I & T \end{bmatrix}$$

$$A \approx \begin{bmatrix} I \\ T_1^{\mathsf{T}} \end{bmatrix} A_{(1:k,1:k)} \begin{bmatrix} I & T_2 \end{bmatrix}.$$

Related but different approaches

- Column/row sampling with leverage scores (Clarkson, Drineas, Kannan, Mahoney, Woodruff, ...)
- CUR decomposition (Tyrtyshnikov, Hackbusch, ...)
- Non-negative factorization (...)
- Nyström interpolation (...)

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Many-body Schrodinger equation

- Consider a quantum system with N_e electrons. The many-body Schrodinger equation for the ground state

$$H\Psi = \left(\sum_{i=1}^{N_e} -\Delta_{x_i} + \sum_{i<j} \frac{1}{|x_i - x_j|} - \sum_{i,\alpha} \frac{M_\alpha}{|x_i - z_\alpha|} \right) \Psi, \quad i\partial_t \Psi = H\Psi$$

  where $\Psi = \Psi(x_1, \ldots, x_{N_e})$ and nuclei at $\{z_\alpha\}$ with charge $\{M_\alpha\}$.

- Ground state: $H\Psi_0 = E_0\Psi_0$
  - E_0 is the lowest eigenvalue. $E_0 = \inf_{\|\Phi\|=1} \langle \Phi | H | \Phi \rangle$.
  - $\Psi_0 = \Psi_0(x_1, \ldots, x_{N_e})$ is the lowest eigenfunction.
- Density $\rho(x) = N_e \int |\Psi_0(x, x_2, \ldots, x_{N_e})^2| dx_{2,\ldots,N_e}$.
- High-dimensional problem and curse of dimensionality.

Kohn-Sham DFT

$$H\Psi = \left(\sum_{i=1}^{N_e} -\Delta_{x_i} + \sum_{i<j} \frac{1}{|x_i - x_j|} - \sum_{i,\alpha} \frac{M_\alpha}{|x_i - z_\alpha|} \right) \Psi, \quad i\partial_t \Psi = H\Psi$$

- Kohn-Sham density function theory: 3D nonlinear problem

$$(-\Delta + V[\rho])\,\psi_i = \lambda_i \psi_i, \quad i = 1, \ldots, N_e.$$

$\lambda_1, \ldots, \lambda_{N_e}$ are the smallest N_e eigenvalues.
$\psi_1, \ldots, \psi_{N_e}$ are the Kohn-Sham (KS) orbitals (eigenfunctions)

$$\rho(x) = \sum_{i=1}^{N_e} |\psi_i(x)|^2.$$

- Nonlinear eigenvalue problem: self consistent iteration

$$\rho(x) \Rightarrow V[\rho](x) \Rightarrow \{\psi_i(x)\}_{1 \le i \le N_e} \Rightarrow \rho(x) \Rightarrow V[\rho](x) \Rightarrow \ldots$$

- At the end, we hold converged $\{\psi_i(x)\}_{1 \le i \le N_e}$ and $\rho(x)$.

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Kohn-Sham orbitals and subspace

$$\begin{cases} (-\Delta + V[\rho])\,\psi_i = \lambda_i \psi_i, \quad i = 1, \ldots, N_e \\ \rho(x) = \sum_{i=1}^{N_e} |\psi_i(x)|^2. \end{cases}$$

$$\rho(x) \Rightarrow V[\rho](x) \Rightarrow \{\psi_i(x)\}_{1 \le i \le N_e} \Rightarrow \rho(x) \Rightarrow V[\rho](x) \Rightarrow \ldots$$

- This is the starting point of computing other physical quantities.
- Many such quantities depend only on the subspace spanned by $\{\psi_i(x)\}_{1 \le i \le N_e}$.

  e.g. $\rho = \mathrm{diag}(\Psi\Psi^{\mathsf{T}}), \quad \Psi = \begin{bmatrix} \psi_1 & \ldots & \psi_{N_e} \end{bmatrix}, \quad \Psi\Psi^{\mathsf{T}} = \text{proj. op.}$

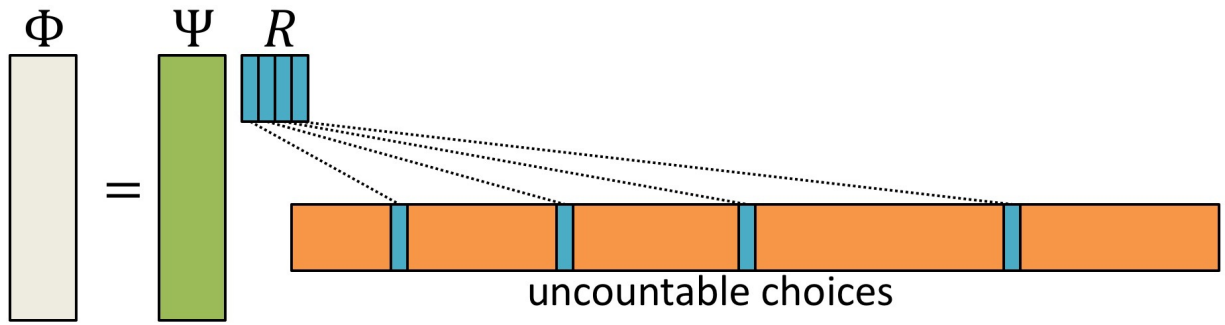
- Seek for a sparse and localized basis for $\mathrm{span}\,\Psi$
  - Interpretability.
  - Computation and storage efficiency.

Localization [with A. Damle and L. Lin]

- Given $\Psi = [\psi_1, \ldots, \psi_{N_e}] \in \mathbb{R}^{n \times N_e}$, find $R \in \mathbb{R}^{N_e \times N_e}$ such that

$$\Phi = [\phi_1, \ldots, \phi_{N_e}] = \Psi R$$

has localized and well-conditioned columns.



- Assumption: working with insulators so such a basis exists.
- Requirements
  - Sparse/localized.
  - Orthogonal (at least well-conditioned).

Previous work

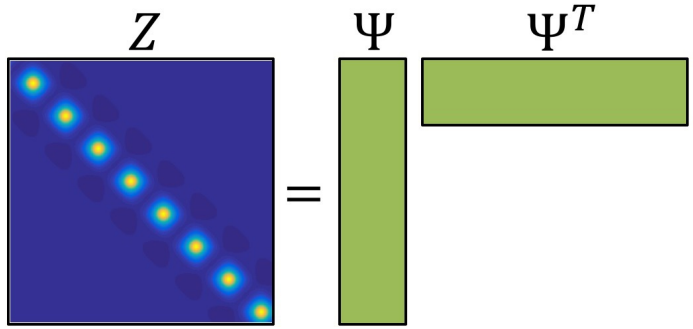
- Maximally localized Wannier functions [Marzari+Vanderbilt].

$$\min_{R \in \mathbb{SO}_k} \sum_{i=1}^{N_e} \left(\int r^2 |\phi_i^R(r)|^2 dr - \left(\int r |\phi_i^R(r)|^2 dr \right)^2 \right).$$

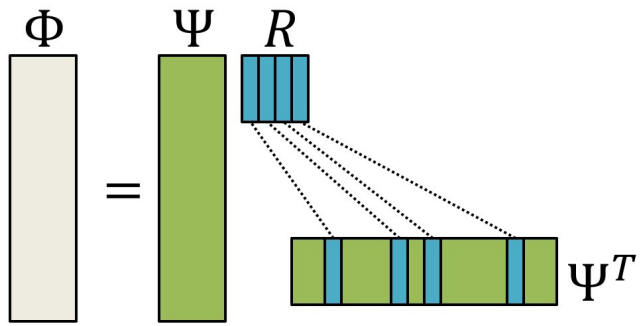
- Challenges
  - Non-convex optimization problem.
  - Needs smart initial guess.

Density matrix

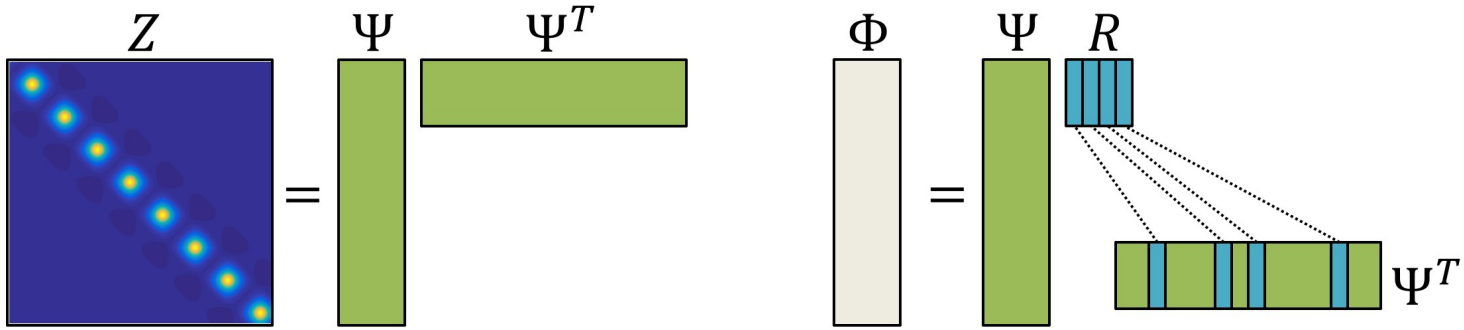
- Requirement 1: sparse/localized
- Idea: consider the density matrix (projector) $Z = \Psi\Psi^{\mathsf{T}}$.
- For insulators, Z has localized and sparse columns.



- Instead of using arbitrary columns for R, only look for selected columns of the density matrix (SCDM).



Interpolative decomposition



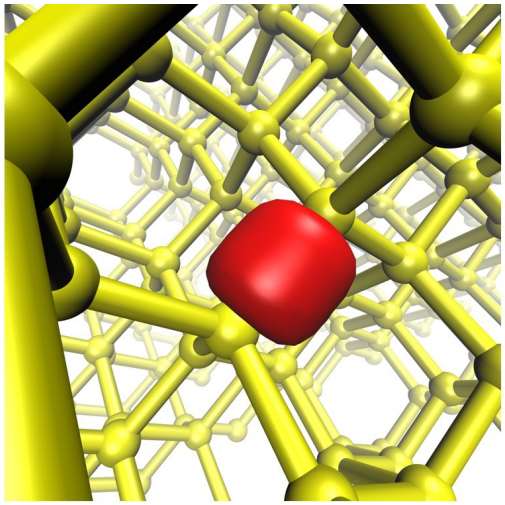
- Requirement 2: orthogonal or at least well-conditioned
- Apply ID to Ψ^{T}: (let C be the picked columns)

$$R := (\Psi^{\mathsf{T}})_{(:,C)}, \quad \Phi = \Psi R = \Psi(\Psi^{\mathsf{T}})_{(:,C)}.$$

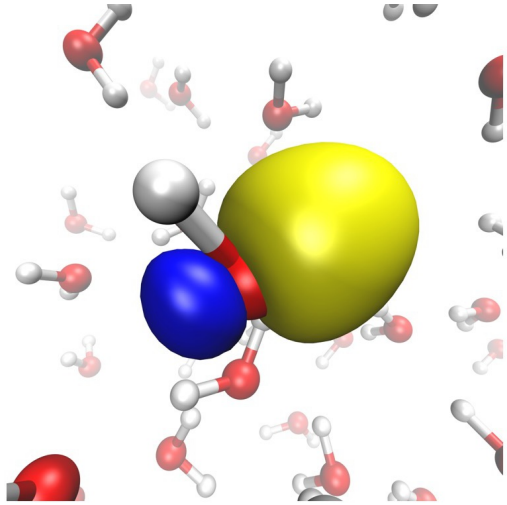
- For orthonormal Φ, the QRCP gives $\Psi^{\mathsf{T}} P = Q \begin{bmatrix} R_{11} & R_{12} \end{bmatrix}$:

$$\Phi = \Psi Q.$$

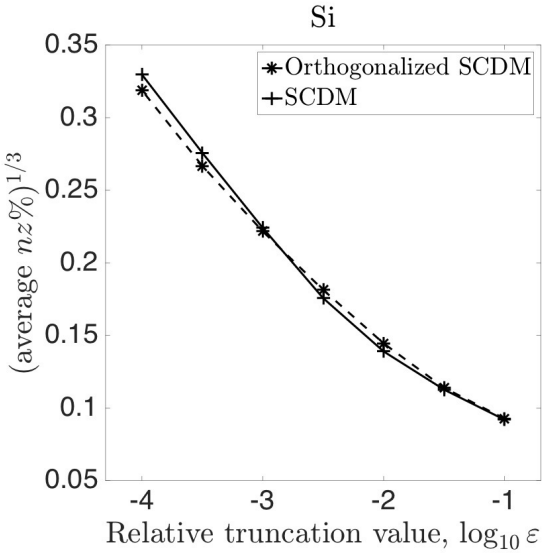
SCDM example: silicon and water

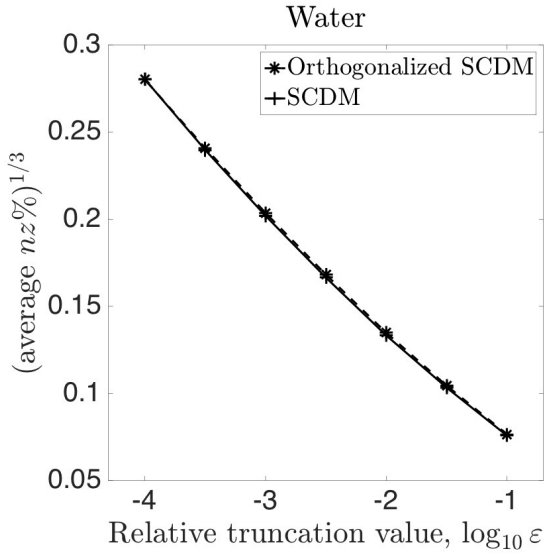


Yellow: Silicon crystal structure
Red: isosurface of localized orbital



Red and white: water molecule structure. Blue and yellow: isosurface of localized orbital





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Electron repulsion integral tensor [with J. Lu]

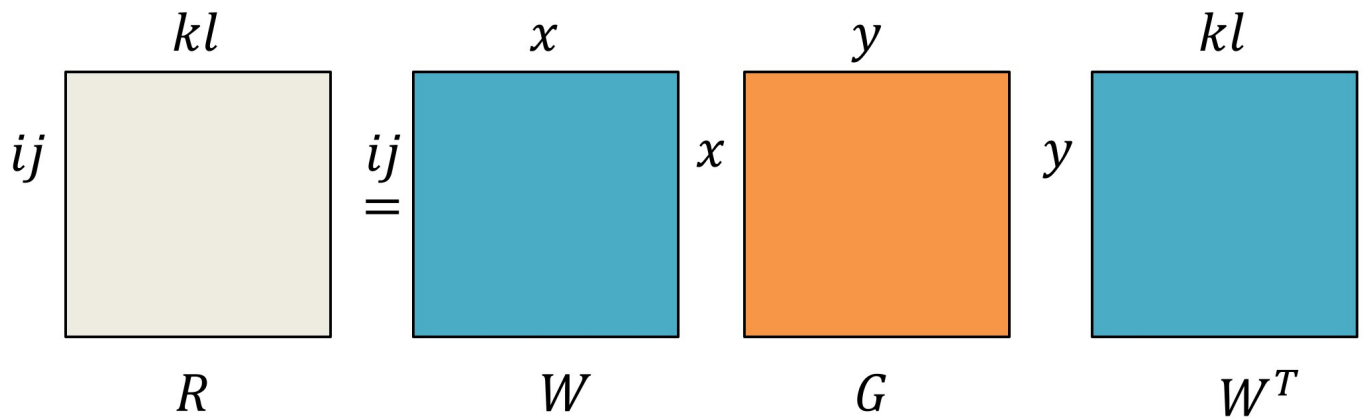
- Given a set $\{\psi_i(x)\}_{1\le i\le N_e}$ of basis functions.
- Electron repulsion integral tensor $R \in \mathbb{R}^{N_e^2 \times N_e^2}$

$$R_{ij,kl} = \iint \psi_i(x)\psi_j(x)\frac{1}{|x-y|}\psi_k(y)\psi_l(y)dxdy.$$

(e.g. $N_e = 10^3$ and $n = 10^6$).

- Introduce $W \in \mathbb{R}^{N_e^2 \times n}$ and $G \in \mathbb{R}^{n\times n}$ with $W_{ij,x} = \psi_i(x)\psi_j(x)$ and $G_{x,y} = \frac{1}{|x-y|}$

$$R = WGW^{\mathsf{T}}.$$



- Goal: compute and represent $R = (R_{ij,kl})$ efficiently.

Interpolative separable density fitting (ISDF)

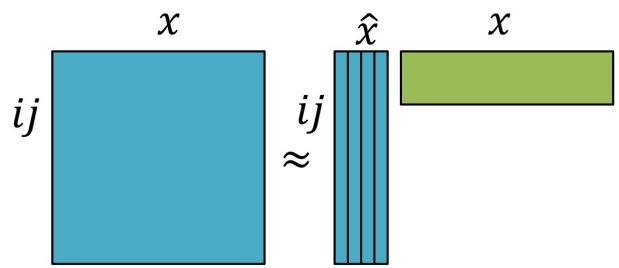
- Each column of W is separable (i.e. an outer-product)

$$W_{ij,x} = \psi_i(x)\psi_j(x)$$

- In most cases W is numerically low-rank.
- Consider $\psi_i(x) = \exp\left(2\pi\sqrt{-1}ix\right)$,

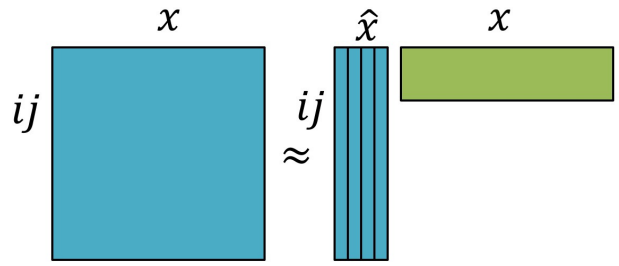
$$\psi_i(x)\psi_j(x) = \exp\left(2\pi\sqrt{-1}(i+j)x\right).$$

- Only $O(N_e)$ choices for $(i+j)$ for N_e^2 combinations of i and j.
- Idea: W has rank $O(N_e)$. Apply ID to compress W.

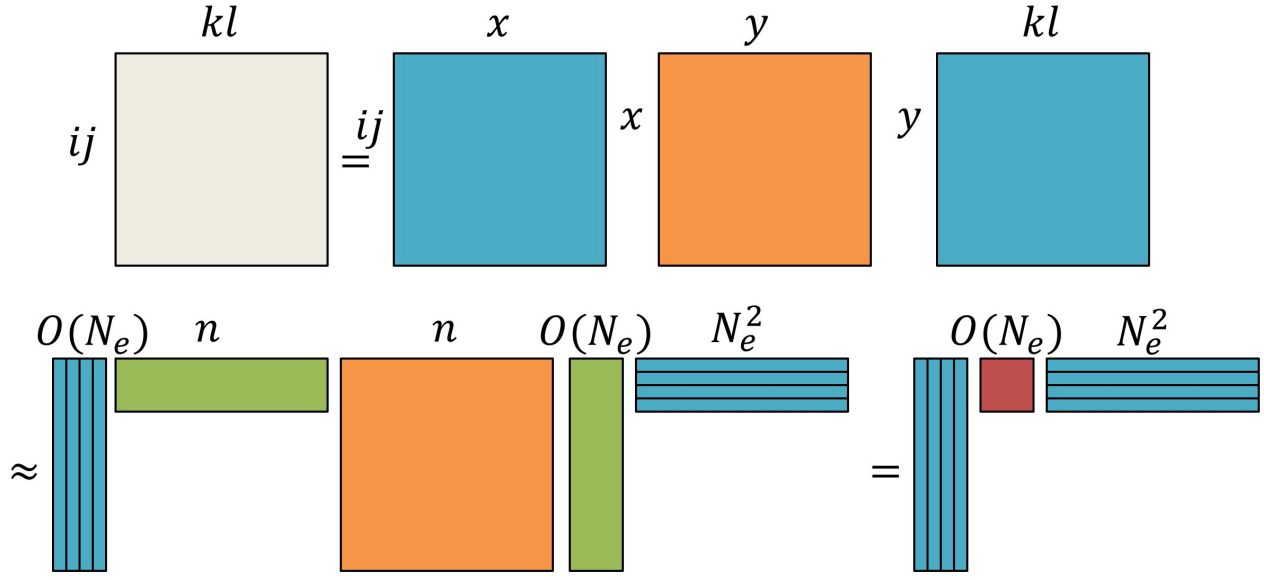


Interpolative separable density fitting (ISDF)

- Given an interpolative decomposition of W



- Then $R = WGW^{\mathsf{T}}$ has approximation

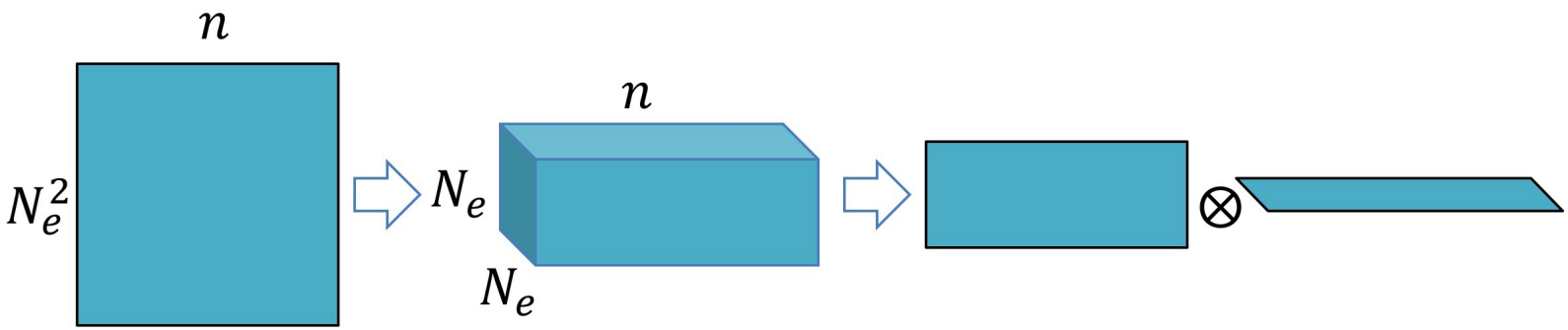


Middle matrix products done with FFTs.

- Costs: $O(N_e^2)$ storage and $O(N_e^2 n)$ time

How to compute the ID of W

- Size of W: $N_e^2 \times n$
- Naive randomized ID costs at lest $O(N_e^3 n)$
- Idea: use the separable (outer-product) structure of W's columns
- Reshape W from $N_e^2 \times n$ to $N_e \times N_e \times n$



- Randomized ID with random project in each dimension of size N_e
- Cost: $O(N_e^2 n)$ time

- So total cost of ISDF: $O(N_e^2)$ storage and $O(N_e^2 n \log n)$ time

Thank you

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- Research supported by NSF and DOE