Operator calculus of density matrices and sparse wavelet representation

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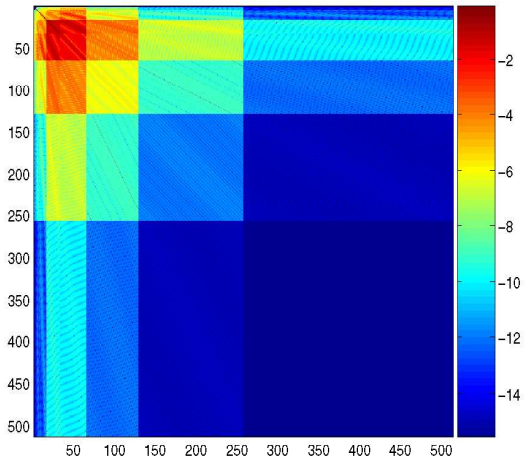

Efficiency

Minimize number of basis functions +
minimize computational complexity

$\Leftrightarrow$

Maximize the accuracy

in electronic structure calculation

Analysis of convergence rates,
development of error estimators,
employment of adaptivity and multigrid methods etc.

Best N-Term Approximation

Consider a Riesz (quasi-orthogonal) basis $\Psi = (\psi_\lambda)_{\lambda \in I}$ in some Hilbert space H.
For $v \in \mathrm{H}$, we consider the **error** of the **best n-term approximation**

$$\sigma_{n,\mathrm{H},\Psi}(v) = \min\{\|v - \sum_{\lambda \in \mathcal{T}} w_\lambda \psi_\lambda\| : w_\lambda \in \mathbb{C}\ ,\ \mathcal{T} \subset I\ ,\ \sharp\mathcal{T} = n\}.$$

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$$\mathrm{A}^s(\mathrm{H},\Psi) := \{v \in \mathrm{H} : \sigma_{n,\mathrm{H},\Psi}(v) \lesssim n^{-s}\} \qquad \text{with} \qquad \|v\|_{\mathrm{A}^s} := \sup_{n \in \mathbb{N}} n^s \sigma_{n,\mathrm{H},\Psi}(v) + \|v\|$$

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Approximation strategy: Keep the n largest coefficients w_λ in $v = \sum_{\lambda \in I} w_\lambda \psi_\lambda$.

Hilbert space: $\mathrm{H} = L^2$ or $\mathrm{H} = H^1$, space of functions in L^2 with bounded kinetic energy $\langle Tv, v\rangle < \infty$.

Basis functions

Candidates:

- Canonical orbitals (=eigen functions of the Fock operator), not known a priori
- Local orbitals (Wannier functions), not known a priori
- atomic type orbitals: Slater type orbitals, Gaussian type orbitals
- ”systematic” basis functions: plane waves, finite elements, multiscale basis functions

Model problem

- effective one particle model (Hartree-Fock, density functional theory, hybrid models):

$$\mathcal{H}_\rho = -\frac{1}{2}\Delta + \mathcal{V}_{core} + \mathcal{V}_{pseudo} + \mathcal{V}_H(n) + \alpha\mathcal{V}_{XC}(n) - \frac{1}{2}\beta\mathcal{W}(\rho),$$

where

$$\rho(\mathbf{x},\mathbf{y}) = \sum_{i=1}^{N/2} \varphi_i(\mathbf{x})\overline{\varphi_i(\mathbf{y})} \qquad (\textbf{density matrix}), \qquad n(\mathbf{x}) = 2\rho(\mathbf{x},\mathbf{x}) \qquad (\textbf{electron density})$$

- aim: linear scaling methods
  - Linear scaling methods using Gaussian type basis functions are limited to ”nondiffusive” basis functions. Furthermore, scaling with respect to the number of basis functions is not linear!
  - Competing with discretizations like plane waves or Finite Differences, which do not (easily) admit linear scaling approaches.

Multiscale Analysis

Practical and/or analytical tool

Multiscale hierarchy: $V_j = \text{span}\{\phi_{j,k} : k \in \Delta_j\}$

$$(V_{-l} \subset V_{-l+1} \subset \ldots) V_0 \subset V_1 \subset \ldots \subset V_j \subset V_{j+1} \subset \ldots \subset L^2$$

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Multiscale decomposition and wavelets

- Decomposition: $V_{j+1} = V_j \oplus W_j, \qquad V_J = \bigoplus_{j=-1}^{J-1} W_j, \qquad W_{-1} := V_0$
- Wavelets: $W_j = \operatorname{span}\{\psi_{j,k} : k \in \nabla_j := \Delta_{j+1} \setminus \Delta_j\} = \operatorname{span} \Psi_j$
- local supports: $\operatorname{diam} \operatorname{supp} \psi_{j,k} \sim 2^{-j} \;\; , \;\; \operatorname{diam} \operatorname{supp} \phi_{j,k} \sim 2^{-j}$
- Orthonormality: $\langle \psi_{j,k}, \psi_{j',k'} \rangle = \delta_{(j,k),(j',k')}$
- *d* vanishing moments: $|\langle \psi_{j,k}, x^\alpha \rangle| = 0 \;\; , \;\; |\alpha| < d.$

Theorem (Norm equivalences for Sobolev spaces):

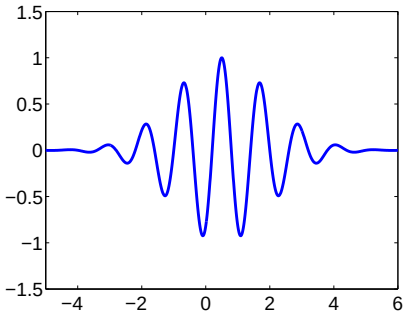
For all $0 \leq s < \sup\{s \in \mathbb{R} : \phi \in H^s(\mathbb{R})\}$ there holds ($\lambda = (j,k)$, $|\lambda| = j$)

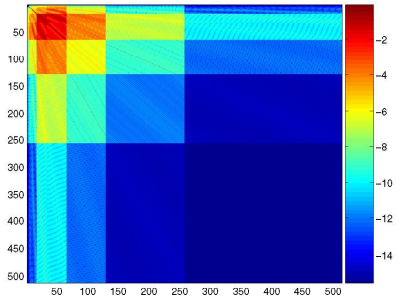
$$\|u\|_s^2 \sim \sum_{\lambda} |\langle u, \psi_\lambda \rangle|^2 2^{2|\lambda| s}$$

Sparse grid - hyperbolic cross

$$\psi_{\mathbf{j},\mathbf{k}}(\mathbf{x}) = \psi_{j_1,k_1}(x_1) \cdot \ldots \cdot \psi_{j_N,k_N}(x_N) \ , \ \Psi = \Psi \otimes \ldots \otimes \Psi = \bigotimes_{i+1}^{N} \Psi \ ,$$

$$\|\mathbf{j}\|_1 = \sum_{i=1}^{N} j_i \leq L \ , \ \sharp\Psi \sim L^{N-1} 2^L \sim n \log^{N-1} \text{ instead of } n^N \ !$$



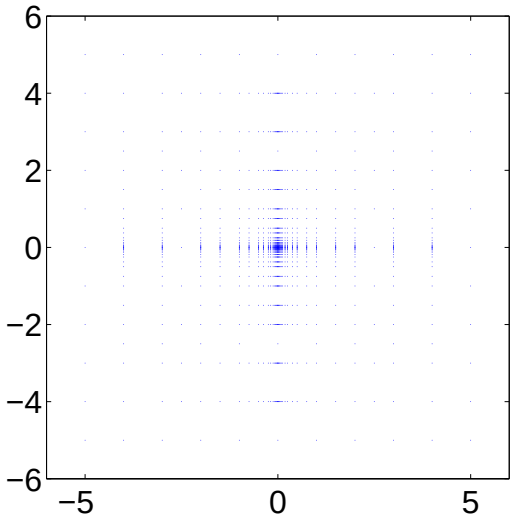


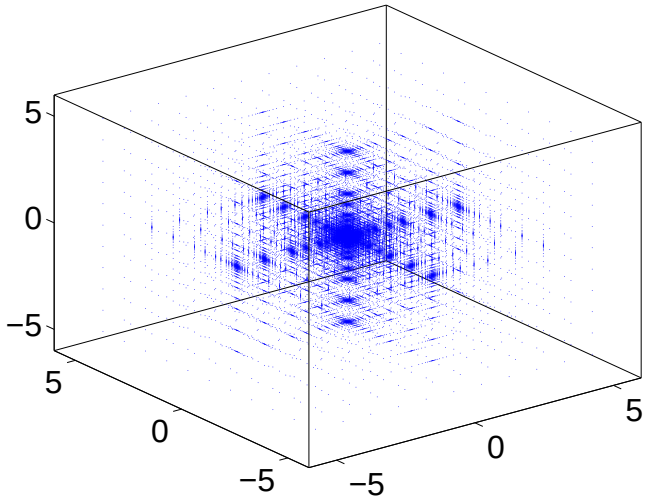
Theorem (Flad, Hackbusch, Schneider '05) :

- For every $0 < s < d - 1$ (d vanishing moments) the singularity Φ of the electron-nuclear cusp and therefore the canonical orbitals (Lieb-Simon) φ_i belong to $\mathrm{A}^s(H^1(\mathbb{R}^3), \bigotimes_{i=1}^{3} \Psi)$, (instead of $\mathrm{A}^{s/3}$, but GTO bases are exponentially convergent).

- For every $0 < s < d - 1$ the density matrix ρ belongs to $\mathrm{A}^s(H^1(\mathbb{R}^3) \otimes H^1(\mathbb{R}^3), \bigotimes_{i=1}^{6} \Psi)$, (instead of $\mathrm{A}^{s/6}$).

Adaptive sparse grid H-atom:





Nonlocal Operators – Wavelet Matrix Compression

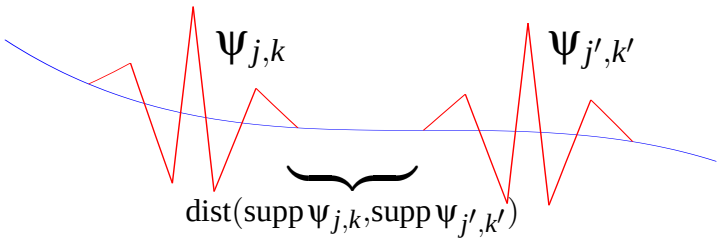
Operators $\mathcal{A} : H^t(\mathbb{R}^3) \to H^{-t}(\mathbb{R}^3)$ with a Schwartz kernel satisfying a Calderón Zygmund type estimate

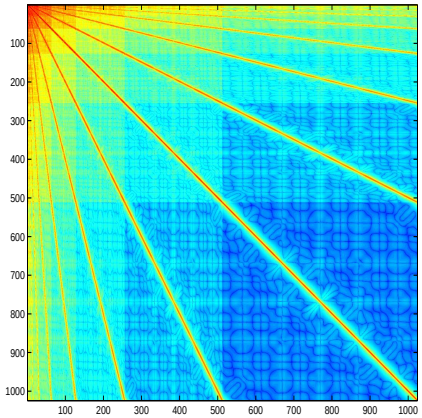
$$|\partial_{\mathbf{x}}^{\alpha}\partial_{\mathbf{y}}^{\beta}K(\mathbf{x},\mathbf{y})| \lesssim \frac{1}{|\mathbf{x}-\mathbf{y}|^{3+2t+|\alpha|+|\beta|}} \quad , \quad \mathbf{x} \neq \mathbf{y} \, ,$$

have matrix coefficients

$$|\langle \mathcal{A}\psi_{j,k}, \psi_{j',k'}\rangle| \leq c \, \frac{2^{(j+j')(\tilde{d}+3/2)}}{\operatorname{dist}(\operatorname{supp}\psi_{j,k}, \operatorname{supp}\psi_{j',k'})^{3+2t+2\tilde{d}}}$$

1. Compression:
(Dahmen-Prößdorf-Schneider,
von Petersdorff-Schwab)





$\Rightarrow$ with these arguments we can proof that, for distant pairs the energy contribution in MP2 calculation decays

$$O(R^{-6})$$

Basic observation:

$$\langle \varphi_a \varphi_j | r_{1,2}^{-1} | \varphi_b \varphi_l \rangle \lesssim R^{-3}$$

Theorem (Flad, Hackbusch, Schneider '05) :

- **Linear scaling complexity**: The density matrix ρ, and the ground state energy of an effective single particle model (HF, LDA, etc.), can be computed with an accuracy $\varepsilon \leq n^{-s}$, $s < d-1$, within (log-)linear complexity $O(n(\log^2 n))$.

- For every $0 < s < 1/2$ the singularity $\Theta \approx |\mathbf{x}-\mathbf{y}|$ of an electron-electron cusp (Kato, Hoffmann-Ostenhoff et al.) belongs to $\mathrm{A}^s(H^1(\mathbb{R}^6), \Psi_x \otimes \Psi_y)$, (instead of $\mathrm{A}^{s/2}$, but the same as for GTO bases).

- **Claim**: The wave function Ψ belongs to $\mathrm{A}^s(H^1(\mathbb{R}^{3N}), \bigotimes_{i=1}^{N} \Psi)$, $0 < s < \frac{1}{2}$.

Kronecker tensor product

- Separation of variables:

$$F(x_1,\ldots,x_N,y_1,\ldots,y_N) \approx \sum_{k=1}^{p} f_{1,k}(x_1,y_1)\cdot\ldots\cdot f_{N,k}(x_N,y_N)$$

- Kronecker product decomposition of tensors:

$$\mathbf{A} = \sum_{k=1}^{p} \mathbf{U}_{1,k}\otimes\ldots\otimes\mathbf{U}_{N,k}$$

- Important to improve the computation of

$$(\langle \mathcal{V}\psi_\lambda,\psi_{\lambda'}\rangle)$$

- Rank one updates (Grasedyck, Chinnamsetty et al. '04). See **Poster**

- Expanding $|\mathbf{x}-\mathbf{y}|$ (Hackbusch '04)

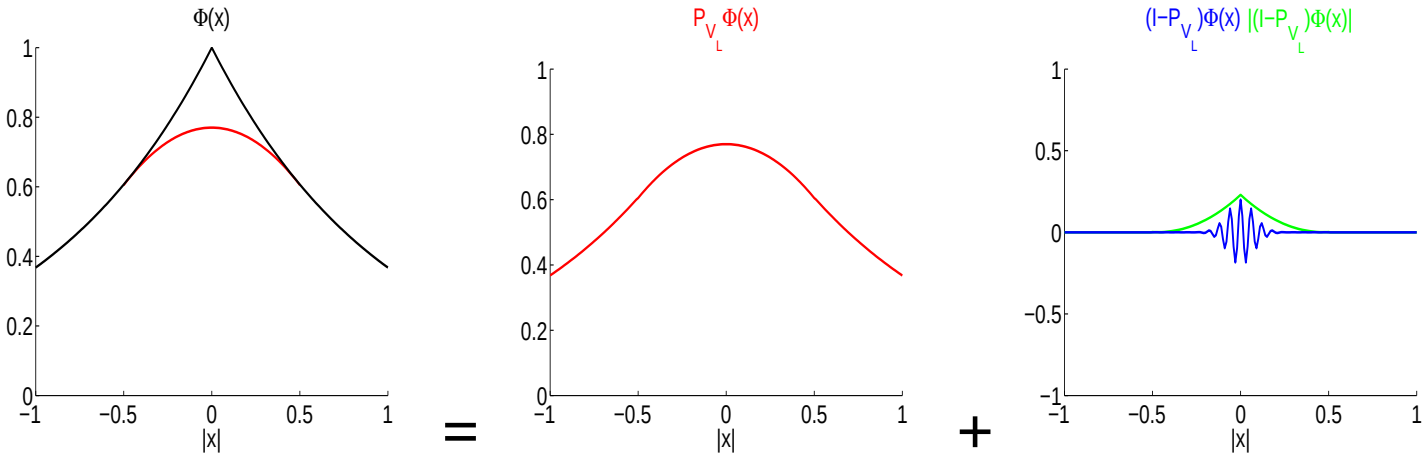
Contracted basis functions and separation of scales

- split orbital Φ into relatively smooth part $P_{V_L}\Phi$ and singular part $(I - P_{V_L})\Phi$:

$$\Phi = P_{V_L}\Phi + (I - P_{V_L})\Phi, \qquad P_{V_L}\Phi \perp (I - P_{V_L})\Phi$$

- molecular scale: smooth part $P_{V_L}\Phi$ by mesh widths $h = 2^{-j} \approx 0.2 \ldots 0.5$

- atomic scale: approximate singular part by contractions of higher-level basis functions with small support – by the projections $(I - P_{V_L})\Phi_{STO/GTO}$ of the core singularities (oszillating and only few per atom)



Error decay rates for the ground state energy

- Multiscale bases and sparse grids: n degrees of freedom

Hyperbolic cross	(Yserentant '04, '05)	$O(n^{-1/3})$
Adaptive sparse grids	(H., F., S.)	$O(n^{-s})$ for all $s < 1$
		$O(n^{-2})$ using $r_{1,2}$ singularity functions
		Demand: linear complexity $O(n)$

- GTO bases: basis set of size $\mathcal{N}$ and n degrees of freedom

CCSD : $n \sim (N^2(\mathcal{N}-N)^2) \sim \mathcal{N}^2$	$O(\mathcal{N}^{-1}) = O(n^{-1/2})$
$r_{1,2}$-Methods CCSD	$O(\mathcal{N}^{-2}) = O(n^{-1})$
Linear scaling CCSD: $n \sim \mathcal{N}$	$O(\mathcal{N}^{-1}) = O(n^{-1})$

Actually complexity is worse.

- Monte-Carlo methods: n sample points $\quad O(n^{-1/2})$