A Reconstruction Approach for Electronic Structure Computation

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Acknowledgments

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Outline

- Overview of Relevant DFT-OFDFT Material
- Proposed Method
- Preliminary Numerical Results
- Current & Future Work
- Concluding Remarks
The DFT avenue to Electronic Structure Computation

- Hohenberg and Kohn (1964)
  - For N-representable electron densities:
    - The electron density uniquely defines the state of the system
    - A system has an electronic density distribution that minimizes its energy

- Kohn-Sham method
  - Scales like $O(N^3)$
  - Methods have been proposed that scale like $O(N)$
The OFDFT flavor of DFT

- Componentwise, energy functional assumes form
  
  \[ E[\rho; \{R_A\}] = T[\rho] + K[\rho] + J[\rho] + E_{ne}[\rho; \{R_A\}] \]

- Getting correct kinetic energy functionals
  - Elusive task, very much work in progress
  - Conceptually, we aimed at a density independent kernel

\[ T[\rho] = T_{TF}[\rho] + T_{vW}[\rho] + T_{Hyb}[\rho] \]

\[ T_{TF}[\rho] = C_{TF} \int dr \rho^3(r) \]

\[ T_{vW}[\rho] = \frac{1}{8} \int dr \frac{| \nabla \rho(r)^2 |}{\rho(r)} \]

\[ T_{Hyb}[\rho] = \sum_\alpha T_\alpha[\rho] \]

\[ T_\alpha[\rho] = \int \int dr \, dr' \rho^\alpha(r) \rho^\alpha(r') w_\alpha(r - r') \]
OFDFT

- Thumbs up:
  - No orbital localization
  - No orthonormalization
  - No Brillouin-zone sampling
  - Scales like $O(N)$

- Thumbs down
  - Getting right expression for KE remains very much a challenge
  - Model transmisibility is low

- Goal of this work
  - It is not to advance OFDFT analytical foundation…
  - Rather, leverage computational power and applied math to extend size of tractable problems treated ab-initio


Goal

- Develop real-space method that reduces the dimension of the problem
  - High resolution where needed
  - Divide and conquer approach for parallel computation:
    - Each subdomain has a dedicated process
  - Where feasible, only use a subset of the subdomains and use reconstruction to obtain the density in the excluded subdomains
    - Equivalent to model reduction ("gaptooth" idea)
Basic Idea

- Reduce the number of “degrees of freedom” in the energy minimization problem
  - Use an interpolation operator to express the discounted degrees of freedom

- Example: **Electronic Structure Computation** - only degrees of freedom from $D_0, D_1, Y_1, Y_2, Y_3, D_{11},$ and $D_{12}$ are considered in the problem

- Additionally, only $Y_1, Y_2, Y_3$ are used to recover (**through interpolation**) the value of the electronic density in $D_3, D_4, D_5, D_7, D_8, D_9$
Solution Approach

- Mesh the entire domain of investigation $D$

- Partition the domain $D$ in active and passive (reconstructed) subdomains

- Use grid-based discretization of energy functional to express energy $E$ exclusively in terms of density $\hat{\rho}$
  - $\hat{\rho}$ represents value of charge density measured at the grid points of reconstruction subdomains

- Solve optimization problem
  - Adjust value of density at grid points of reconstruction subdomains to minimize the electronic energy functional
**Fundamental Idea**

- Need to evaluate integrals of the form
  \[ I(\Theta[\rho(r)]) = \int \Theta[\rho(r)] \, dr \]

- An assumption is made in regards to the form of \( \Theta \):
  \[ \Theta[\rho(r)] = L(r) \cdot R[\rho(r)] \]

- Example:
  \[ I(\Theta[\rho(r)]) = E_{ne} = -\int \rho(r) \sum_{A=1}^{M} \frac{Z_A}{|r - \mathbf{R}_A|} \, dr \quad \Rightarrow \quad \begin{aligned} R[\rho(r)] &= \rho(r) \\ L(r) &= \sum_{A=1}^{M} \frac{-Z_A}{|r - \mathbf{R}_A|} \end{aligned} \]

- **Two essential steps:**
  - Use grid values and quadrature rules to evaluate integral
  - Use reconstruction approach to approximate \( R[\rho(r)] \) in passive subdomains
OF-DFT Energy Evaluation

- Integrals are evaluated using a quadrature rule:

\[
I[\Theta] = \int L(r) R[\rho(r)] dr = \sum_{i=1}^{n} \left[ \sum_{k \in Q(i)} w_{i,k} L(r_{i,k}) R(r_{i,k}) \right]
\]

- Reconstruction idea:

\[
R(r_{i,k}) = \sum_{\alpha=1}^{p} \nu_{i,\alpha} R_{i,\alpha}
\]

- Elementary manipulations lead to matrix representation:

\[
I[\Theta] = \mathcal{K}^T[L] R[\hat{\rho}]
\]

- \(\mathcal{K}[L]\) - Kernel vector (constant, evaluated at beginning of simulation)
- \(R[\hat{\rho}]\) - Vector based on values of \(\hat{\rho}\) at grid points of reconstruction subdomains
Evaluating the TF Energy Functionals

- **Kinetic Energy** (kernel $\mathcal{K}$)
  \[
  T[\rho(r)] = C_F \int \rho^5(r) \, dr = C_F \mathcal{K}^T \rho^5 \quad (L(r) = 1)
  \]

- **Exchange correlation** (kernel $\mathcal{K}$)
  \[
  K[\rho(r)] = -C_X \int \rho^4(r) \, dr = -C_X \mathcal{K}^T \rho^4 \quad (L(r) = 1)
  \]

- **Electron-Nuclei interaction** (kernel $\mathcal{K}_{en}$)
  \[
  E_{en}[\rho, \{R_A\}] = -\sum_{A=1}^{M} \int \frac{Z_A \rho(r)}{||R_A - r||} \, dr = \mathcal{K}_{en}^T \rho \quad \left( L(r) = \sum_{A=1}^{M} \frac{-Z_A}{||r - R_A||} \right)
  \]

- **Electron-Electron interaction** (kernel $\mathbf{K}$)
  \[
  J[\rho] = \frac{1}{2} \int \int \frac{\rho(r) \rho(r')}{||r - r'||} \, dr \, dr' = \frac{1}{2} \mathbf{\rho}^T \mathbf{K} \mathbf{\rho} \quad \left( L(r', r) = \frac{1}{||r' - r||} \right)
  \]
Double Integral Kernel $K$

- Computation of $K$ is expensive
- Computation of $K$ done in *parallel*
  - Scales extremely well (parallel divide and conquer)
- Kernel $K$ stays constant, computation done *once*

$$K = K_1 + K_2 + K_3 + K_4 + K_5$$
Solving the Optimization Problem

- Solver used: TAO – parallel optimization solver
  - Bound-constrained optimization: $\hat{\rho}$ should stay positive
  - Penalty approach, due to charge conservation constraint

Optimization Problem:

$$
\min E = C_F \mathbf{K}^T \hat{\rho}^5 - C_X \mathbf{K}^T \hat{\rho}^3 + \mathbf{K}^T \hat{\rho} + \frac{1}{2} \hat{\rho}^T \mathbf{K} \hat{\rho}
$$

s.t. \(0 = \mathbf{K}^T \hat{\rho} - N\)

- $\hat{\rho}$ represents the value of the density at the grid points

- Variable mesh is essential
Numerical Results (1)

- DFT: Thomas-Fermi-Dirac representation

- 1D problem with 11 nuclei of H
  - Problem partitioned in 11 subdomains
  - 3 reconstruction subdomains
  - 6 passive (reconstructed) subdomains
  - 50 nodes per cell

- Problem implemented in AMPL, an off-the-shelf optimization package
Reconstruction Error

Relative Error between the direct and interpolation reconstruction method
Numerical Results: 3D Simulations

- Parallel function/gradient evaluation
- Parallel optimization solver

- 5 parallel processes
- 5 active subdomains

- 5 parallel processes
- 3 active subdomains
Numerical Results: 3D Simulations

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Numerical Results: 3D Simulations

- 5 parallel processes
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Future Plans

- Better DFT and OFDFT
  - LDA
  - More sophisticated kinetic energy functionals
    - Can we handle density dependent kernels for KE?
  - Pseudopotentials

- Parallel Computers: provide speed and storage

- Apply to some real problems (monoatomic metals)
Conclusions

- Model reduction idea applied to OFDFT leads to smaller optimization problem

- Early simulation results obtained on parallel architectures
  - Reconstruction approach leverages
    - Parallel function evaluation
    - Parallel bound constraint optimization solver (TAO)

- A whole more left to be done, it is very much work in progress