# A Reconstruction Approach for Electronic Structure Computation

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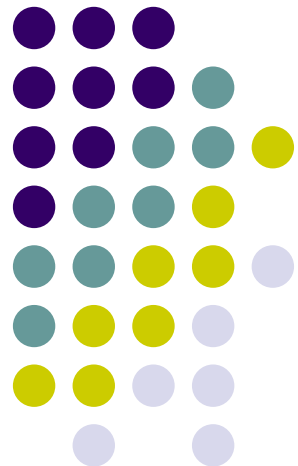


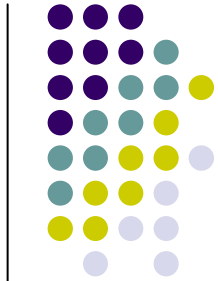
# Acknowledgments


- Collaborators:
  - Dan Negrut (University Wisconsin-Madison)
  - Mihai Anitescu (ANL)
  - Anter El-Azab (Florida State)
  - Steve Benson (ANL)
  - Emil Constantinescu (grad. student, Virginia Tech.)
  - Toby Heyn (undergrad. Student, U Wisconsin

# Outline

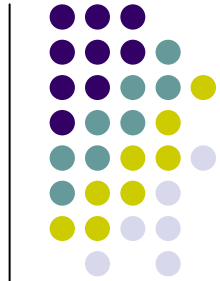
- Overview of Relevant DFT-OFDFT Material
- Proposed Method
- Preliminary Numerical Results
- Current & Future Work
- Concluding Remarks

# 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

$$E\left[\rho;\{\mathbf{R}_A\}\right] = T\left[\rho\right] + K\left[\rho\right] + J\left[\rho\right] + E_{ne}\left[\rho;\{\mathbf{R}_A\}\right]$$

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

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

$$T_{TF}[\rho] = C_{TF}\int \mathrm{d}\mathbf{r}\ \rho^{\frac{5}{3}}(\mathbf{r}) \qquad T_{vW}[\rho] = \frac{1}{8}\int \mathrm{d}\mathbf{r}\ \frac{|\nabla\rho(\mathbf{r})|^2}{\rho(\mathbf{r})}$$

$$\begin{aligned} T_{Hyb}[\rho] &= \sum_\alpha T_\alpha[\rho] \\ T_\alpha[\rho] &= \int\int \mathrm{d}\mathbf{r}\ \mathrm{d}\mathbf{r}'\ \rho^\alpha(\mathbf{r})\rho^\alpha(\mathbf{r}')w_\alpha(\mathbf{r}-\mathbf{r}') \end{aligned}$$

# 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*


- *Radiation Damage*: B. D. Wirth, G. R. Odette, J. Marian, L. Ventelou, J. A. Young-Vandersall, and L. A. Zepeda-Ruiz, J. Nucl. Mater. **329–333**, 103 (2004).

- *Crack Propagation*: C. L. Rountree, R. K. Kalia, E. Lidorikis, A. Nakano, L. Van Brutzel, and P. Vashrishta, Annu. Rev. Mater. Res. **32**, 377 (2004).

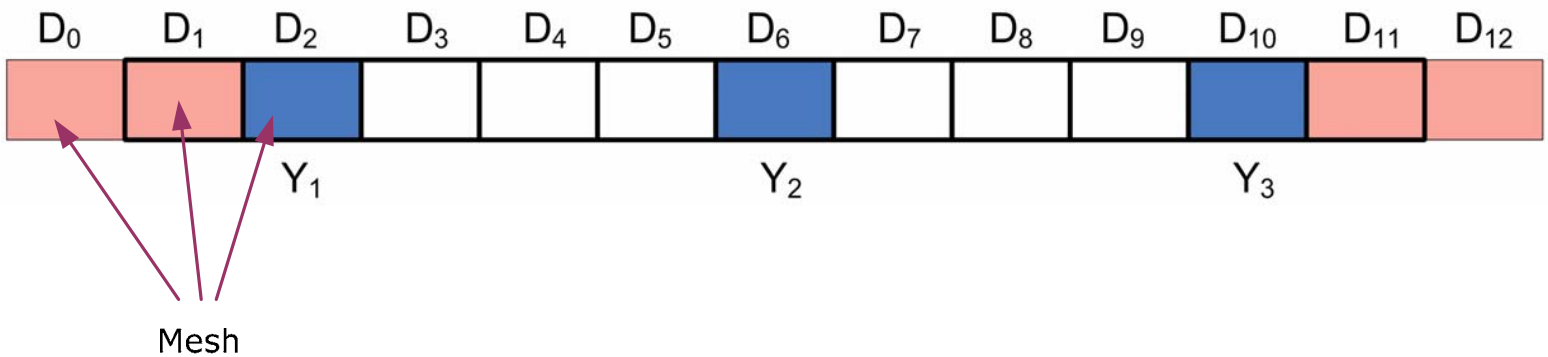
# Goal

- 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

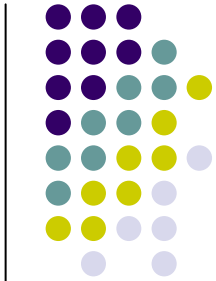
- Reduce the number of “degrees of freedom” in the energy minimization problem
  - Use an interpolation operator to express the discounted degrees of freedom

- 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



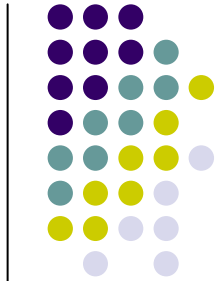

- Additionally, only $Y_1$, $Y_2$, $Y_3$ are used to recover (**through interpolation**) the value of the electronic density in $D_3$, $D_4$, $D_5$, $D_7$, $D_8$, $D_9$

# 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 $\widehat{\rho}$
  - $\widehat{\rho}$ 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

$$I(\Theta[\rho(\mathbf{r})]) = \int \Theta[\rho(\mathbf{r})]\, \mathrm{d}\mathbf{r}$$

- An assumption is made in regards to the form of $\Theta$:

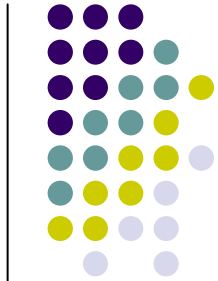
$$\Theta[\rho(\mathbf{r})] = L(\mathbf{r})\, R[\rho(\mathbf{r})]$$

  - Example:

$$I(\Theta[\rho(\mathbf{r})]) = E_{ne} = -\int \rho(\mathbf{r}) \sum_{A=1}^{M} \frac{Z_A}{|\mathbf{r}-\mathbf{R}_A|} \mathrm{d}\mathbf{r} \quad \Rightarrow \quad \begin{cases} R[\rho(\mathbf{r})] = \rho(\mathbf{r}) \\ L(\mathbf{r}) = \sum_{A=1}^{M} \frac{-Z_A}{|\mathbf{r}-\mathbf{R}_A|} \end{cases}$$

- ***Two essential steps***:
  - Use grid values and quadrature rules to evaluate integral
  - Use reconstruction approach to approximate R[ρ(r)] in passive subdomains

# OF-DFT Energy Evaluation



- Integrals are evaluated using a quadrature rule:

$$I[\Theta] = \int L(\mathbf{r})\, R[\rho(\mathbf{r})]\mathrm{d}\mathbf{r} = \sum_{i=1}^{u} \left[ \sum_{k \in Q(i)} w_{i,k}\, L(\mathbf{r}_{i,k})\, R(\mathbf{r}_{i,k}) \right]$$

- Reconstruction idea:

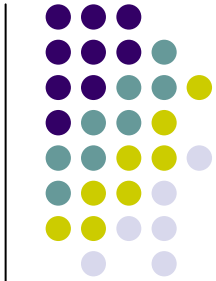
$$R(\mathbf{r}_{i,k}) = \sum_{\alpha=1}^{p} \nu_i^{\alpha} R_{i,k}^{\alpha}$$

- 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 $\mathcal{K}$)

$$T[\rho(\mathbf{r})] = C_F \int \rho^{\frac{5}{3}}(\mathbf{r})\, \mathrm{d}\mathbf{r} = C_F\, \mathcal{K}^T\, \widehat{\rho}^{\frac{5}{3}} \qquad (L(\mathbf{r}) = 1)$$

- Exchange correlation (kernel $\mathcal{K}$)

$$K[\rho(\mathbf{r})] = -C_X \int \rho^{\frac{4}{3}}(\mathbf{r})\, \mathrm{d}\mathbf{r} = -C_X\, \mathcal{K}^T\, \widehat{\rho}^{\frac{4}{3}} \qquad (L(\mathbf{r}) = 1)$$

- Electron-Nuclei interaction (kernel $\mathcal{K}_{\mathrm{en}}$)

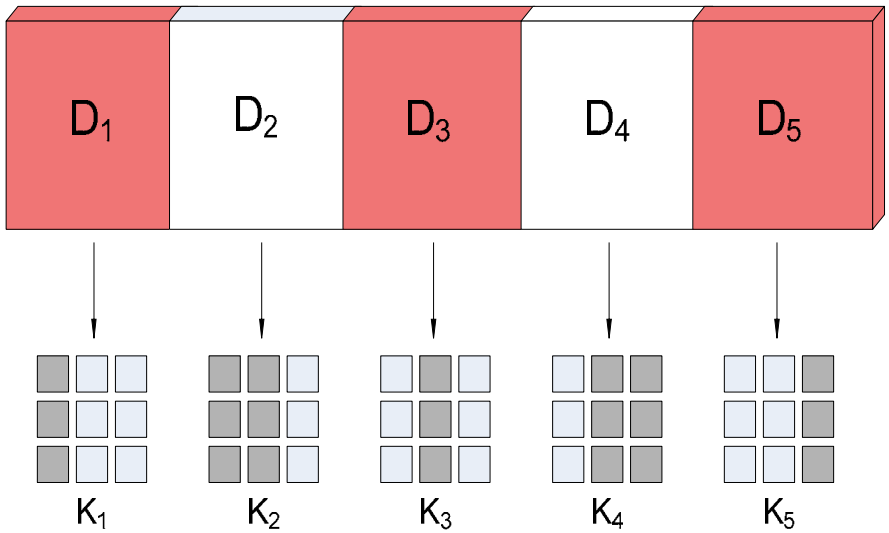
$$E_{en}\,[\rho, \{R_A\}] = -\sum_{A=1}^{M} \int \frac{Z_A\, \rho(\mathbf{r})}{\|\mathbf{R}_A - \mathbf{r}\|}\, \mathrm{d}\mathbf{r} = \mathcal{K}_{en}^T\, \widehat{\rho} \qquad \left( L(\mathbf{r}) = \sum_{A=1}^{M} \frac{-Z_A}{|\mathbf{r} - \mathbf{R}_A|} \right)$$

- Electron-Electron interaction (kernel **K**)

$$J\,[\rho] = \frac{1}{2} \int \int \frac{\rho(\mathbf{r})\, \rho(\mathbf{r}')}{\|\mathbf{r} - \mathbf{r}'\|}\, \mathrm{d}\mathbf{r}\, \mathrm{d}\mathbf{r}' = \frac{1}{2} \widehat{\rho}^T\, \mathbf{K}\, \widehat{\rho} \qquad \left( L(\mathbf{r}', \mathbf{r}) = \frac{1}{|\mathbf{r}' - \mathbf{r}|} \right)$$

# 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*




$$K = K_1 + K_2 + K_3 + K_4 + K_5$$

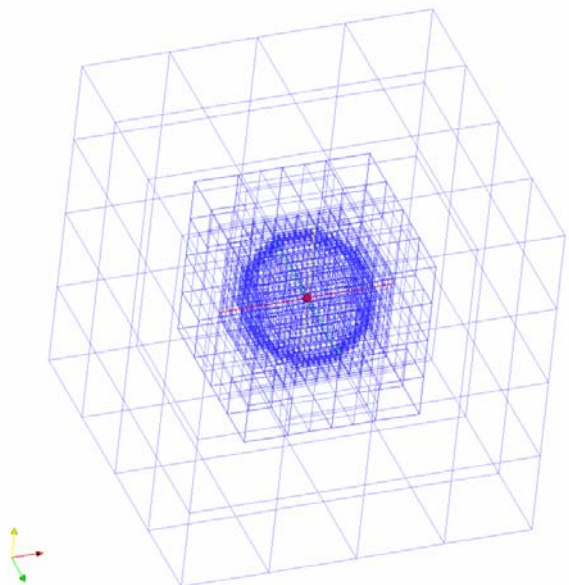
# Solving the Optimization Problem

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

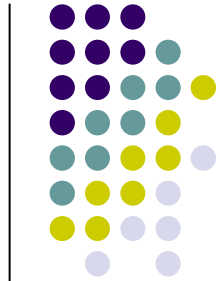
$$\min E = C_F \mathcal{K}^T \hat{\rho}^{\frac{5}{3}} - C_X \mathcal{K}^T \hat{\rho}^{\frac{4}{3}} + \mathcal{K}_{ne}^T \hat{\rho} + \frac{1}{2} \hat{\rho}^T \mathbf{K} \hat{\rho}$$

$$\text{s.t. } 0 = \mathcal{K}^T \hat{\rho} - N$$

- $\hat{\rho}$ represents the value of the density at the grid points
- Variable mesh is essential



# Numerical Results (1)

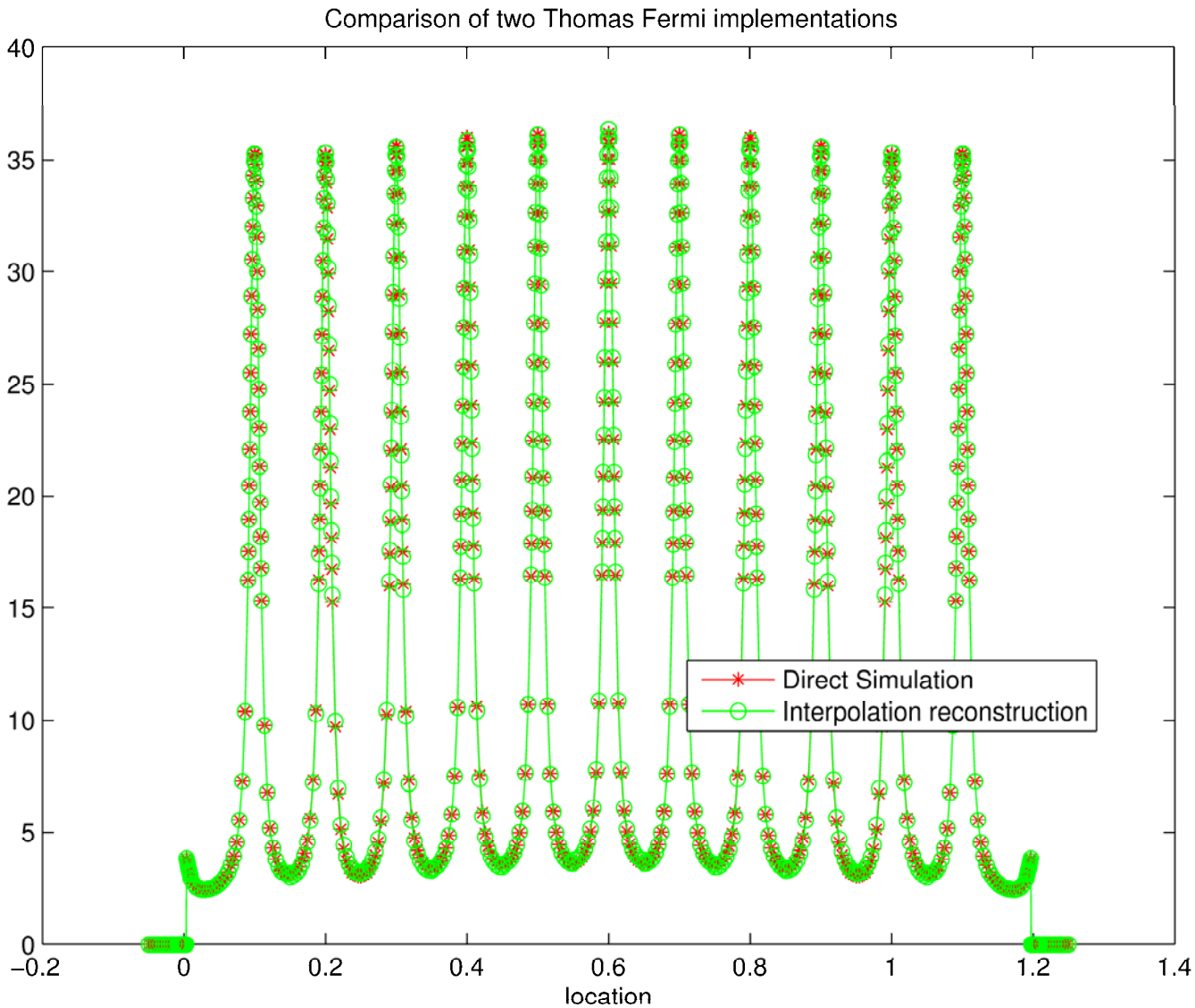


- DFT: Thomas-Fermi-Dirac representation

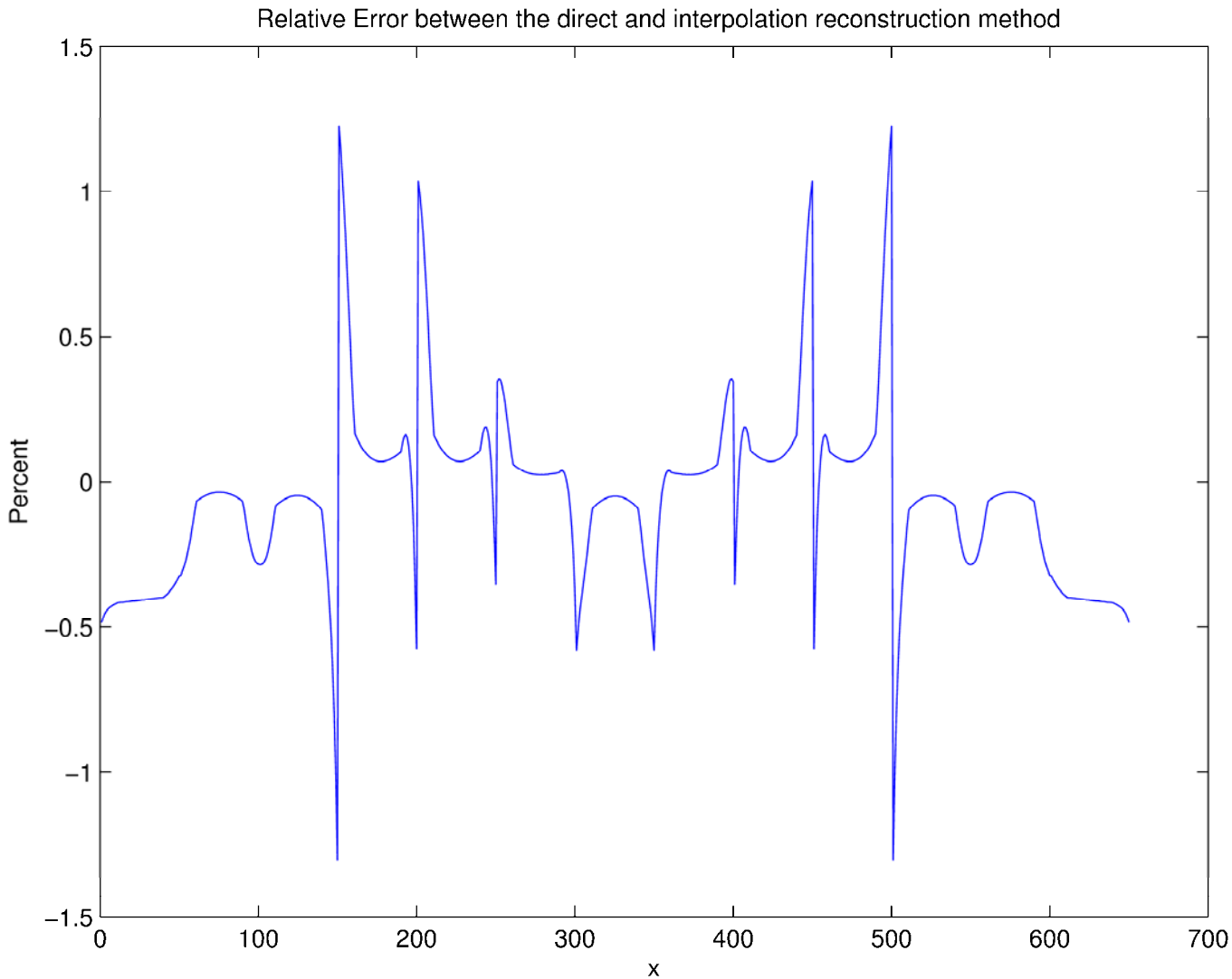
- 1D problem with 11 nuclei of H
  - Problem partitioned in 11 subdomains
  - 3 reconstruction subdomains
  - 6 passive (reconstructed) subdomains
  - 50 nodes per cell

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

# Reconstruction Results



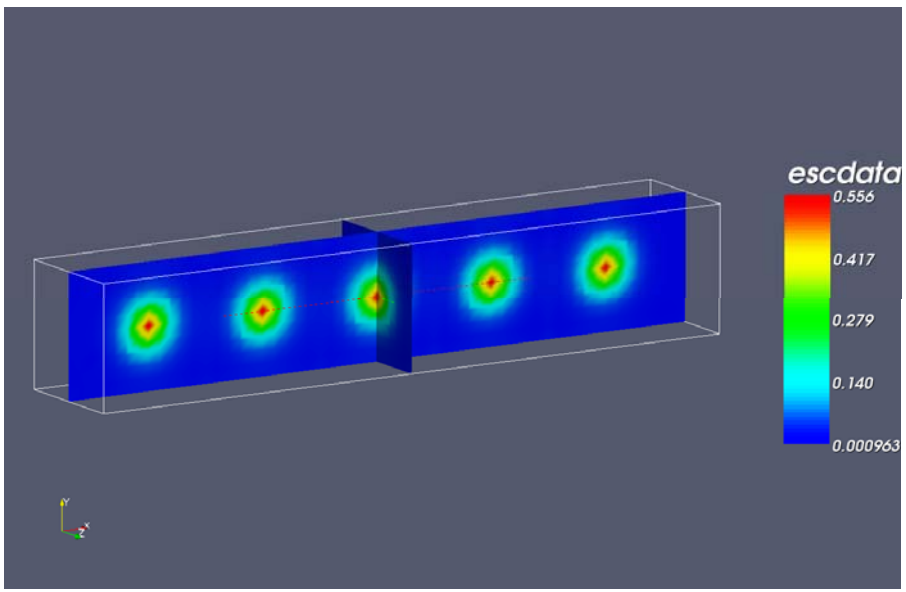
# Reconstruction Error



# Numerical Results: 3D Simulations

- Parallel function/gradient evaluation
- Parallel optimization solver

- 5 parallel processes
- 5 active subdomains

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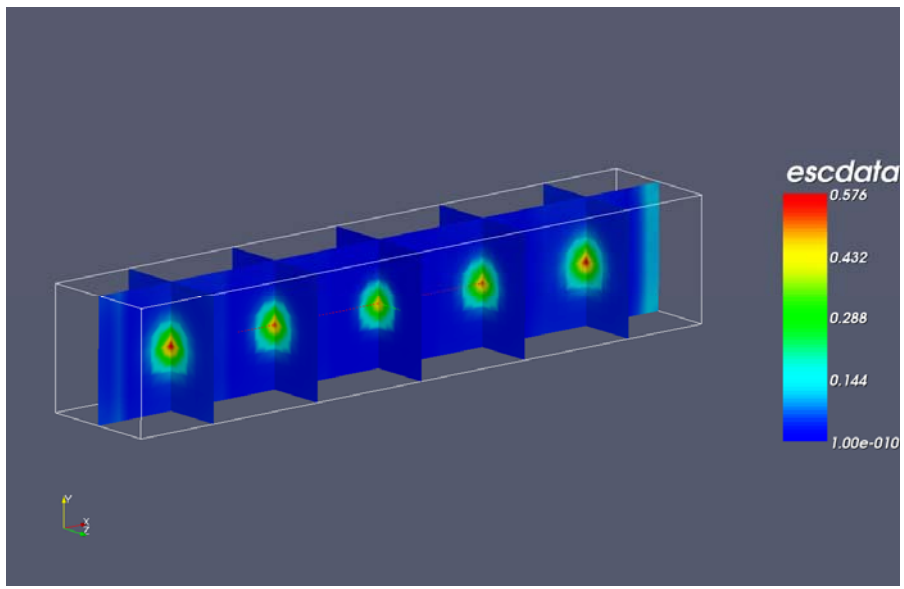

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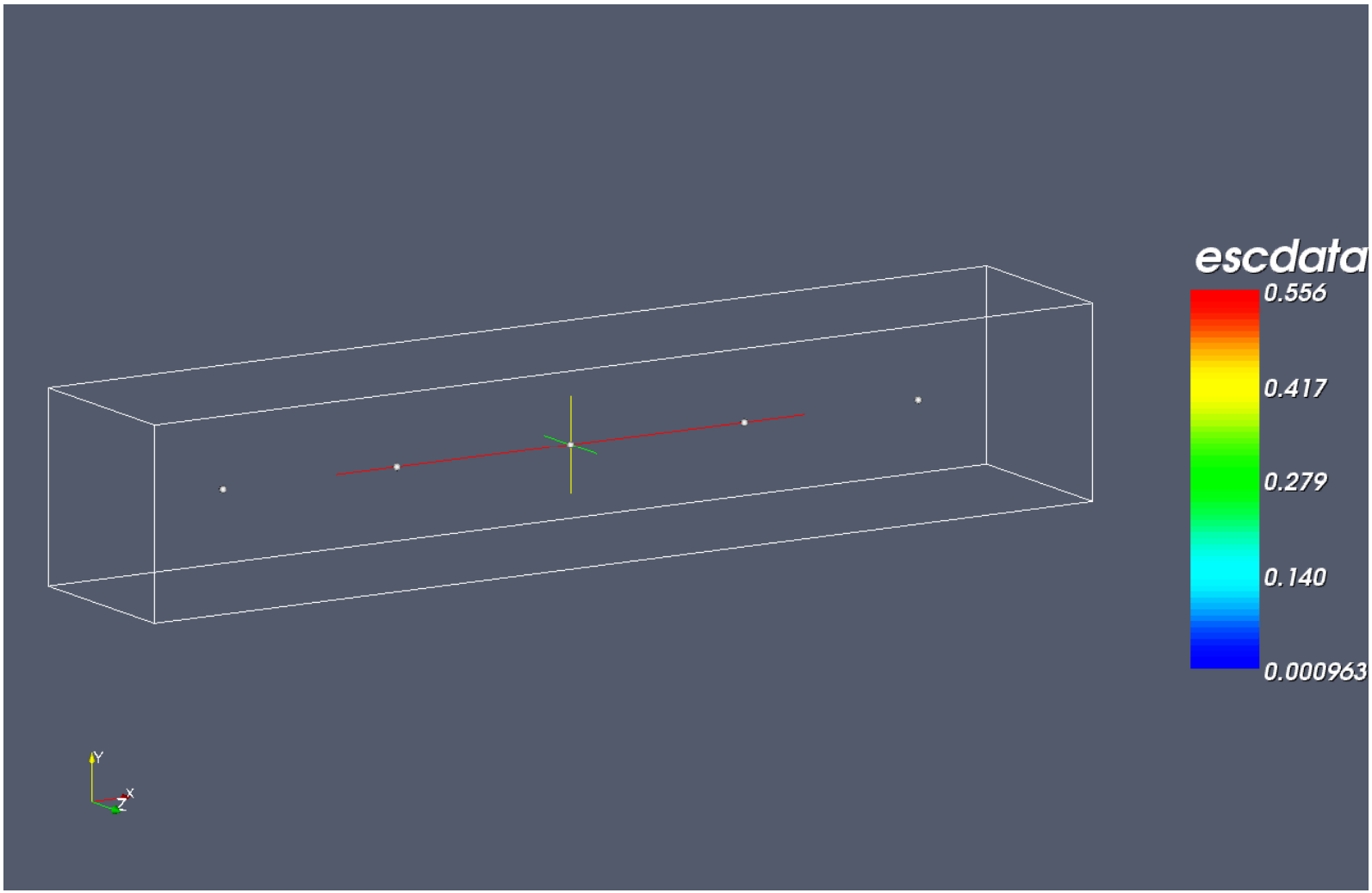

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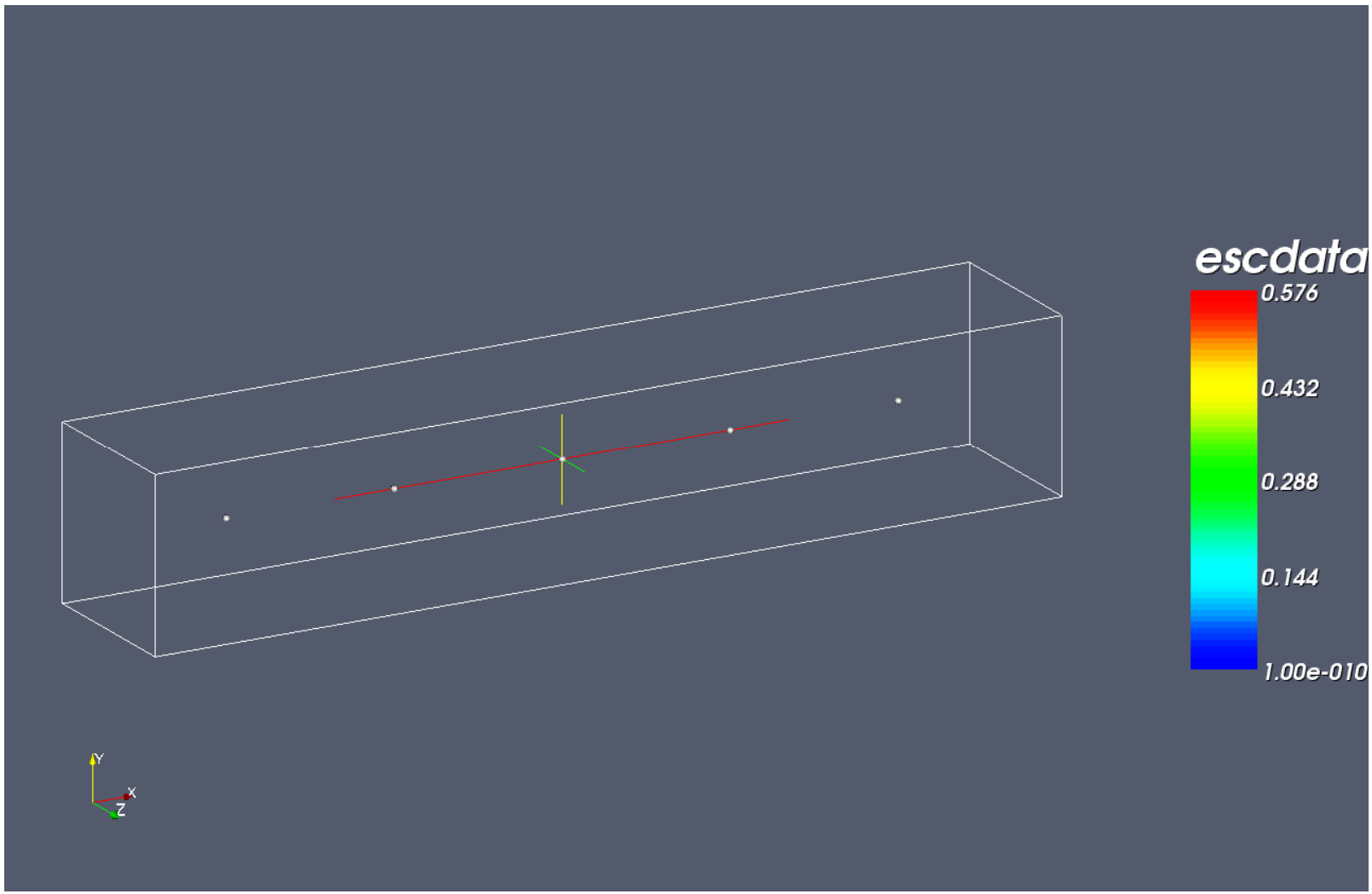
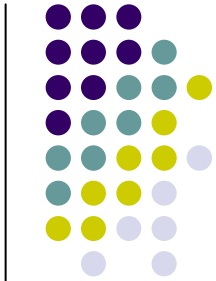


# Future Plans



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