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Investigation of quasicontinuum-like model reduction approaches in material science.

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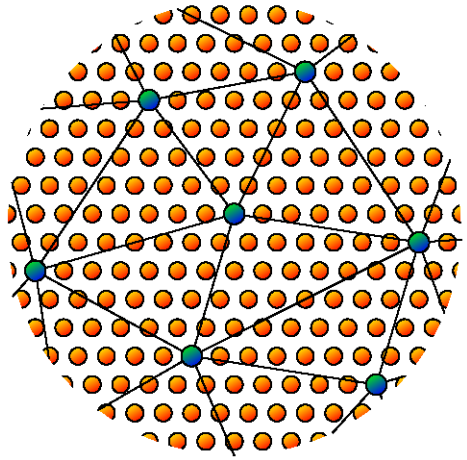
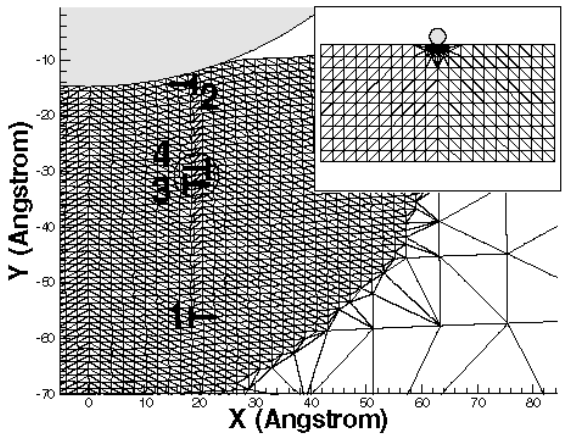
Motivation: Quasicontinuum (QC) Methods in Material Science

- QC: multiscale model reduction method for the simulation of crystalline solids. (Tadmor, Ortiz et a, 96+l)
- In regions of small deformation, positions of “nonrepresentative” atoms are expressed by interpolation of positions of “representative” (local if cut-off) atoms, positioned at nodes of a macro mesh. In “interesting regions” mesh refined to atomic level (“nonlocal”).
- Nanoindentation calculations carried out for ~10^6 atoms with only ~10^4 representative atoms, excellent agreement with full simulations.
- The idea: reduction of degrees of freedom by interpolation

$$x_2 = Tx_1, \dim(x_1) << \dim(x_2)$$





QC mesh and nanoindentation (Tadmor, Philips, et al.)

Motivation: Density Functional Theory

- One of the workhorses of modern computational chemistry.
- The issue is the resolution of the problem (followed by min wrt. $\{R_A\}$)

$$\min_{\widehat{\rho}\geq 0, \int \rho = N} E_{tot}\left[\widehat{\rho}\right] = E_{ne}\left[\widehat{\rho}\right] + J\left[\widehat{\rho}\right] + K\left[\widehat{\rho}\right] + T\left[\widehat{\rho}\right]$$

- Here,

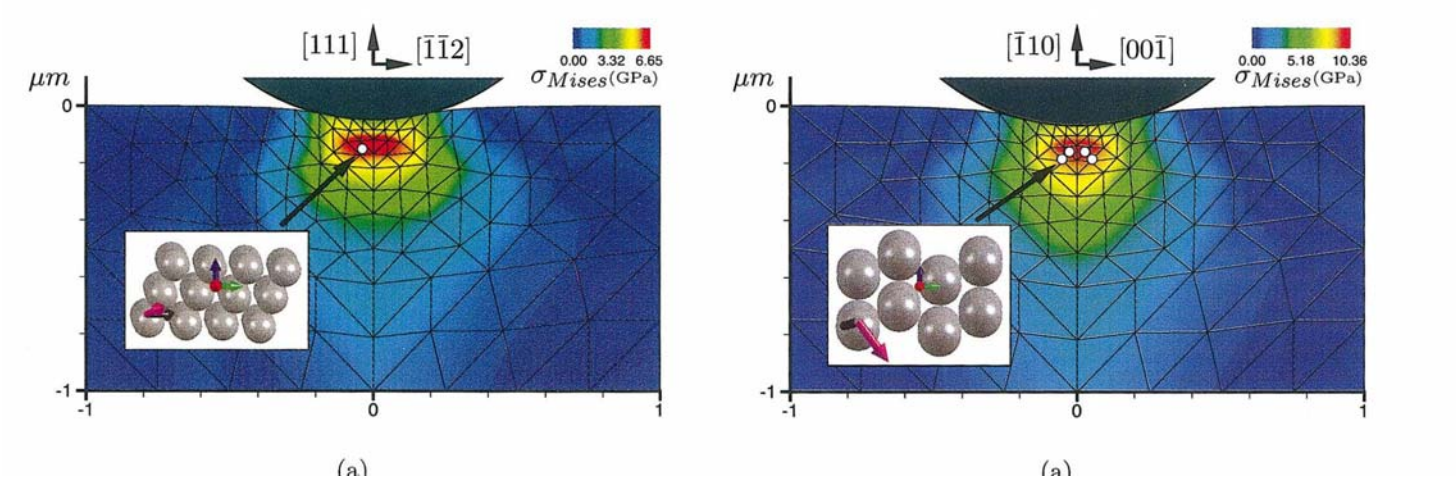
$$J\left[\rho\right] = \frac{1}{2}\int\int \frac{\rho(\mathbf{r})\,\rho(\mathbf{r}')}{\|\mathbf{r}-\mathbf{r}'\|}\,\mathrm{d}\mathbf{r}\,\mathrm{d}\mathbf{r}' \quad E_{ne}(\mathbf{r};\{\mathbf{R}_A\}) = -\sum_{A=1}^{M}\int \frac{Z_A\,\widehat{\rho}(\mathbf{r})}{\|\mathbf{r}-\mathbf{R}_A\|}\,d\mathbf{r}$$

- DFT approaches differ in the way they approximate kinetic energy and exchange energy. In orbital-free (OFDFT) approaches the functionals are explicitly available, not so in the generally more accurate Kohn-Sham.
- Simplest OFDFT Thomas-Fermi. Only for validation of model reduction.

$$T[\rho(\mathbf{r})] = C_F\int \rho^{\frac{5}{3}}(\mathbf{r})\,\mathrm{d}\mathbf{r} \quad K[\rho(\mathbf{r})] = -C_X\int \rho^{\frac{4}{3}}(\mathbf{r})\,\mathrm{d}\mathbf{r}$$

- The main limitation: number of atoms and electrons that can be simulated
- Question: Can a QC-like approach be defined in regions of small deformation and result in model reduction with reasonable accuracy?

Partial answer: DFT based local QC (Fago et al., 04)



- Each representative atom is surrounded by a DFT box.. The electron-nucleus interaction is computed by PBC with the infinite crystal deformed according to the local interpolation rule. (DNS outside reach).
- Problem: the mesh cannot be deformed to the point where DFT boxes interact. So the simulation stops with “initiation of nanoindentation”
- Secondary problem: The problem does not capture the migration of electrons that could accompany such defects.
- Challenge: Move reduction beyond PBC.

Why?

- DFT is necessary: Accurate potential approximation do not exist for many new materials and configurations. But 10^3 atoms all we can do currently (a least with KS need 10^7!)
- The “small deformation of crystal in a large domain” appears in many interesting applications
- Surface effects in nanomaterials, which is essential component of self-assembly.
- Radiation effects/ radiation damage ir materials used in nuclear/fusion reactors. Simulation of primary knock off followed by the “cascade” (Stoller,00) shows “slightly perturbed crystal most places” is a very good hypothesis.

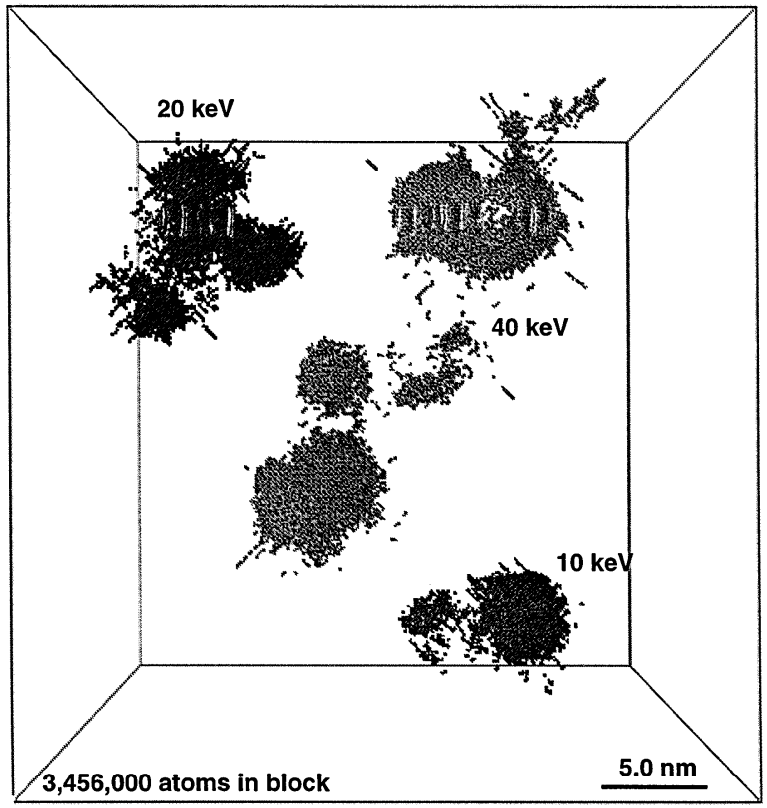


Fig. 4. Illustration of increasing subcascade formation in iron at 100 K as the MD cascade energy increases from 10 to 40 keV.

Outline

- Analysis and variants of interpolation-based (QC like) model reduction in material science.
- A QC-like model reduction approach for orbital free, density functional theory (OFDFT) electronic structure calculations
- Numerical validation of the DFT QC-like model reduction.
- Reasonableness of some of the assumptions.
- Conclusions, Future work.

Colaborators

- Dan Negrut (Wisconsin)
- Anter-El-Azab (Florida State)
- Peter Zapol (Argonne)
- Steve Benson (Argonne).
- Emil Constantinescu (grad stud VPI).
- Toby Heyn (undergrad stud Wisconsin).

Formulation of the problem and the material science approaches

- The problem has a few “representative” degrees of freedom x_1 and a lot of “nonrepresentative” x_2 degrees of freedom – the essence of scale separation.
- Compare with “representative atoms” and “constrained atoms” in “Tadmor et al.”

$$
(\mathrm{O}) \qquad
\begin{array}{lll}
\min_{x_1,x_2} & f(x_1,x_2) & \\
 & g_1(x_1) & = 0 \\
\text{s.t.} & g_2(x_2) & = 0 \\
 & g_3(x_1,x_2) & = 0\,.
\end{array}
$$

The functions $g_1(x_1) : \mathbb{R}^m \longrightarrow \mathbb{R}^{q_1}$, $g_2(x_2) : \mathbb{R}^{n-m} \longrightarrow \mathbb{R}^{q_2}$ and $g(x_1,x_2) : \mathbb{R}^n \longrightarrow \mathbb{R}^{q_3}$ are the constraint functions, which, together with the objective function $f(x_1,x_2) : \mathbb{R}^n \longrightarrow \mathbb{R}$, are twice continuously differentiable.

The two types of reduced problems

- The essential observation is that $x_2 \approx Tx_1$ is a very good approximation for small perturbations of crystalline structure.
- Approach 1: Interpolate and optimize (energy-based)

$$\text{(RO)} \qquad \begin{array}{ll} \min_{x_1} & f(x_1, Tx_1) \\ \text{s.t.} & g_1(x_1) = 0 \\ & g_3(x_1, Tx_1) = 0. \end{array}$$

- Approach 2: Optimize and Interpolate (force-based)

$$\text{(RE)} \qquad \begin{aligned} \nabla_{x_1} f(x_1, Tx_1) + \nabla_{x_1} \langle g_3(x_1, Tx_1), \lambda_3 \rangle \; + & \\ \nabla_{x_1} \langle g_1(x_1, Tx_1), \lambda_1 \rangle \; &= \; 0 \\ g_1(x_1) \; &= \; 0 \\ g_3(x_1, Tx_1) \; &= \; 0. \end{aligned}$$

General Assumptions for Analysis

Interpolation Assumption At the optimal solution $\left(x_1^*, x_2^*\right)$ of the problem (O),

$$\|T\left(x_1^*\right) - x_2^*\| \leq \epsilon,$$

where T is an interpolation operator.

Regularity Assumption The following conditions holds at solution (x^*, λ^*)

· Constraint Qualification Condition (CQC): The rows of the matrices $\nabla_x g_1(x_1)$, $\nabla_x g_2(x_2)$ and $\nabla_x g_3(x_1, x_2)$ are linearly independent.

· Second-Order Sufficient Condition (SOSC):

$$\left.\begin{array}{rcl} \nabla_x g_1(x_1^*)\Delta x &=& 0, \\ \nabla_x g_2(x_2^*)\Delta x &=& 0, \\ \nabla_x g_3(x_1^*, x_2^*)\Delta x &=& 0, \\ \Delta x &\neq& 0 \end{array}\right\} \Rightarrow \Delta x^T \nabla_{xx}^2 L(x^*, \lambda^*)\Delta x > 0.$$

Regularity results for RO problems

- Compatibility conditions for the constraints

$$J_{RO} = \begin{bmatrix} \nabla_{x_1} g_1(x_1^*) \\ \nabla_{x_1} g_3(x_1^*, Tx_1^*) + \nabla_{x_2} g_3(x_1^*, Tx_1^*)T \end{bmatrix} \quad \text{has full row rank}$$

$$g_1(x_1) = 0 \quad \Rightarrow \quad g_2(Tx_1) = 0, \forall x_1$$

. For example: A crystal on a plane surface.

Theorem There exists an ϵ_0 for which, if interpolation assumption is satisfied at $\left(x_1^*, x_2^*\right)$, for $0 \leq \epsilon \leq \epsilon_0$, then the problem (RO) satisfies both the SOSC and the CQC at x_1^* with multiplier $\left(\lambda_1^* + S(x_1^*)^T \lambda_2^*, \lambda_3^*\right)$ and has a solution in a neighborhood of x_1^*.

Further assumptions for (RE) problems

- The proof of regularity of (RE) requires two further assumptions

Assumption (RECF): The constraints of the problem (O) are separable; that is, $g_3 = \emptyset$. Likewise, the constraints $g_2(x_2) = 0$ are linear and satisfy

$$g_1(x_1) = 0 \Rightarrow g_2(Tx_1) = 0.$$

Assumption HT The Hessian of the Lagrangian function satisfies

$$\left\| \nabla^2_{x_2 x_2} L(x^*, \lambda^*) T + \nabla^2_{x_2 x_1} L(x^*, \lambda^*) \right\| \leq \epsilon.$$

Regularity results for (RE) problem

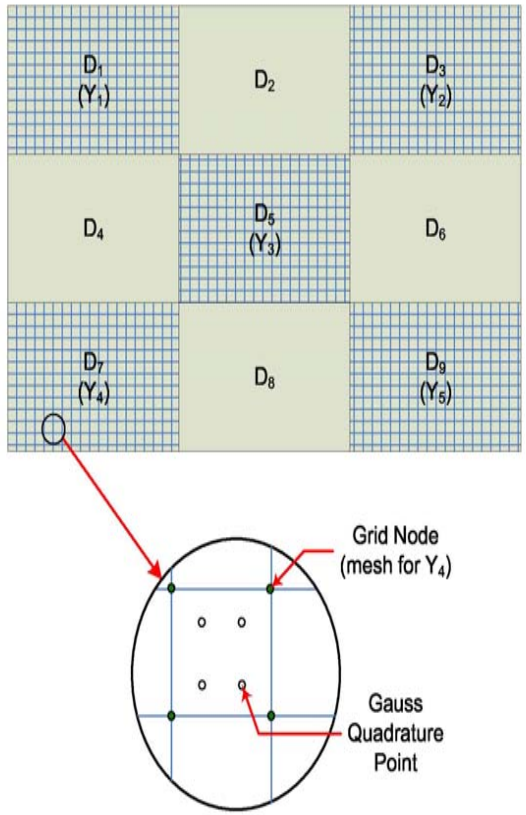
There exists an ϵ_0 for which if the interpolation assumption and HT assumption are satisfied at $\left(x_1^*, x_2^*\right)$, for $0 \leq \epsilon \leq \epsilon_0$, then the problem (RE) has a nonsingular Jacobian at $\left(x_1^*, \lambda_1^*\right)$ as well as a solution in a neighborhood of the same point $\left(x_1^*, \lambda_1^*\right)$.

A multiscale approach for electronic density nanoscale simulations

- Representative variables: The density in the representative domains .
- The interpolation operator is constructed with respect to a reference crystalline mesh

$$Y_\alpha\ ,\ \alpha\ =\ 1,2,\ldots\ ,\ p$$

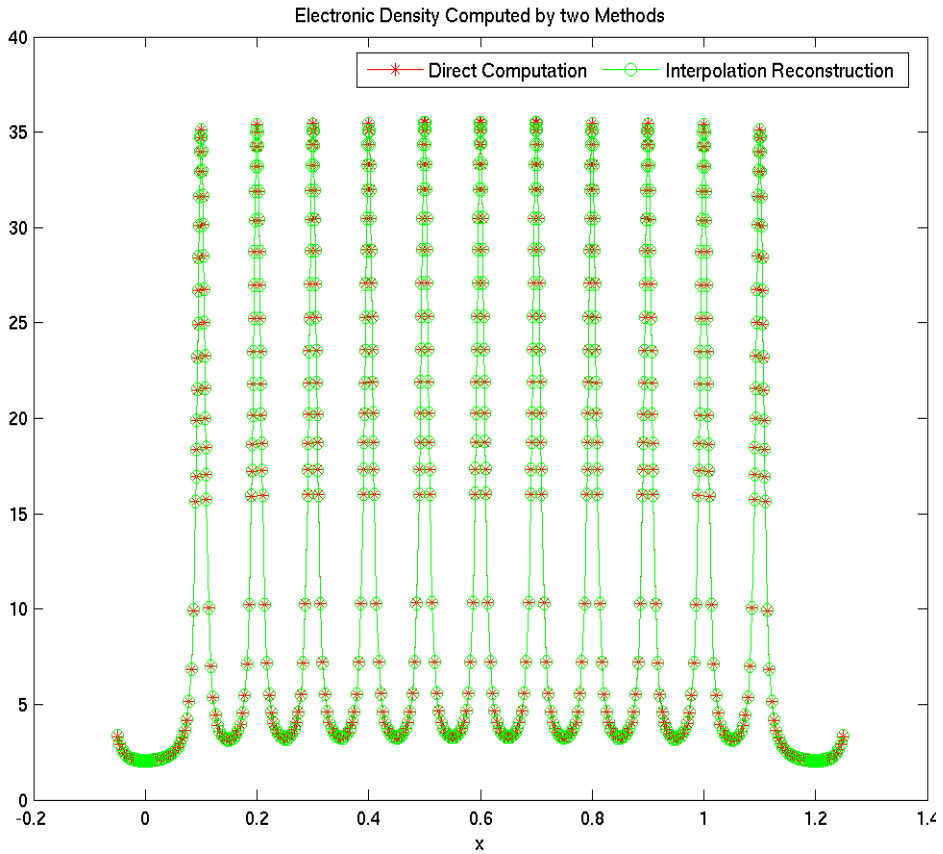
- The approach allows for deformation of the mesh when atoms are also allowed to relax (second part, not covered in our presentation).



$$\rho_i(\Phi(\mathbf{r}^0,t)) = \sum_{\alpha=1}^{p} \vartheta_\alpha(i)\rho_\alpha(\Phi(\mathbf{r}^0+\mathbf{T}_{i\alpha},t)).$$

The nonlinear variational inequality approach

- We substitute the interpolation operator in the optimality conditions
- Example: Thomas-Fermi DFT on 11 Hydrogen atoms, using less than 50% degrees of freedom.
- The bound constraints are nonetheless not active in our example, and we truly have the (RE) approach
- Note that the drift in total charge cannot be captured by PBC.



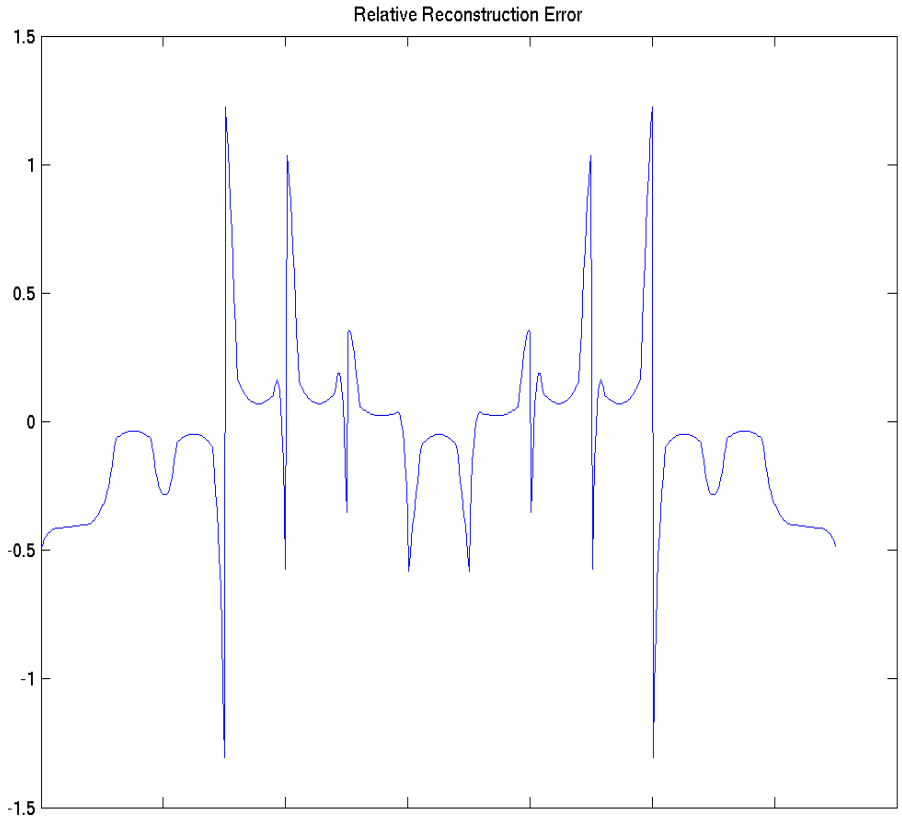
$$0 = g(x_1, Tx_1)$$

$$0 = \nabla_{x_1} f(x_1, Tx_1) + \nabla_{x_1} g(x_1, Tx_1)\lambda.$$

The optimization approach

- Allows us to use optimization tools, with costlier setup but more robustness.
- The maximum relative error is less than 1.5%, remarkable if we consider that we have one order of magnitude variation.

$$\begin{aligned} & \min && f(x_1, Tx_1) \\ (RO)\ & \text{sbj. to} && g(x_1, Tx_1) = 0, \\ & && . \end{aligned}$$



Interpolate and Optimize, one step further

- Interpolation gives assembly rule with precomputable kernels.

$$J(\rho)=\frac{1}{2}\sum_{\alpha=1}^{p}\sum_{\gamma=1}^{p}\int_{Y_\alpha^0}\int_{Y_\gamma^0}\tilde{K}_{\alpha\gamma}(\mathbf{r}^0,\mathbf{r}^{0'})\rho_\alpha(\Phi(\mathbf{r}^0,t))\rho_\gamma(\Phi(\mathbf{r}^{0'},t))d\mathbf{r}^0 d\mathbf{r}^{0'}$$

$$E_{ne}(\rho)=-\sum_{\alpha=1}^{p}\int_{Y_\alpha^0}\tilde{L}_\alpha(\mathbf{r}^0)\rho_\alpha(\Phi(\mathbf{r}^0,t))d\mathbf{r}^0,$$

$$\int\rho d\mathbf{r}=\sum_{\alpha=1}^{p}\int_{Y_\alpha^0}\tilde{M}_\alpha(\mathbf{r}^0)\rho_\alpha(\Phi(\mathbf{r}^0,t))d\mathbf{r}^0.$$

- By a separation of scales argument, we can interpolate, in addition to the state variables, the functionals as well,.(see nect slide)

$$T[\rho]+K[\rho]\approx\sum_{\alpha=1}^{p}\int_{Y_\alpha^0}\tilde{M}_\alpha(\mathbf{r}^0)\theta^1(\rho_\alpha,\Phi(\mathbf{r}^0,t))d\mathbf{r}^0\approx\sum_{\alpha=1}^{p}W_\alpha\left(T[\rho]+K[\rho]\right)_\alpha.$$

Usefulness of further approximations

- Even if the objective function is separable, the "nonrepresentative" part must be explored. Function evaluation still expensive.
- This appears in QC as well, but handled by the fact that the pairwise potential is cut off, and only "nearby" nonrepresentative DOF are explored.
- For many functions, one can accurately interpolate the function values as well, and the same results apply.
- But this must be treated differently for different types of functions (cut-offs dependence) and difficult to formalize.

$$f(x) = f_1(x_1) + f_2(x_2) \Rightarrow$$

$$f_{RO}(x_1) = f_1(x_1) + f_2(Tx_1)$$

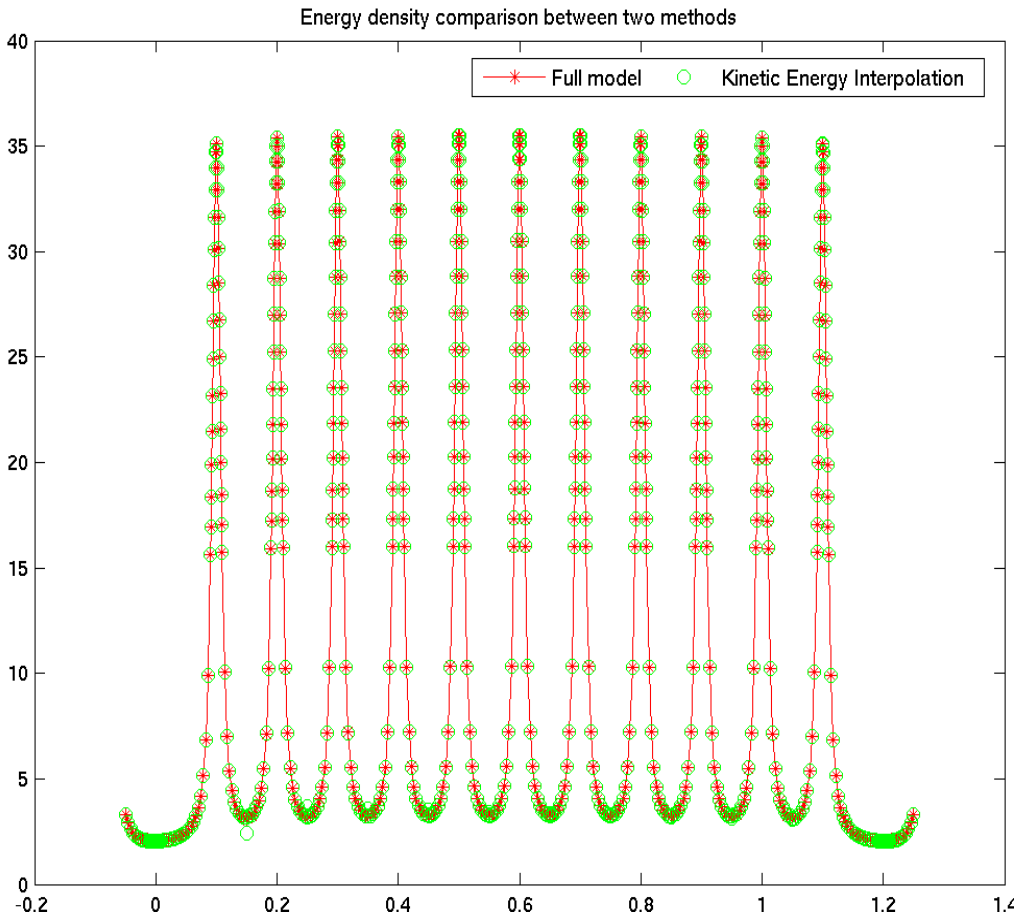
$$f(x) = \sum_{i=1}^{N_{TOT}} f_0(y_i);$$

$$\left(f_0(y_j)\right)_{j \in Nonrep} \approx \left(T\left(\left\{f(y_i)\right\}_{i \in \mathrm{Re}\, pr}\right)\right)_i$$

$$\Rightarrow f_{RO}(x) = \sum_{i \in \mathrm{Re}\, pr} w_i f_0(y_i);$$

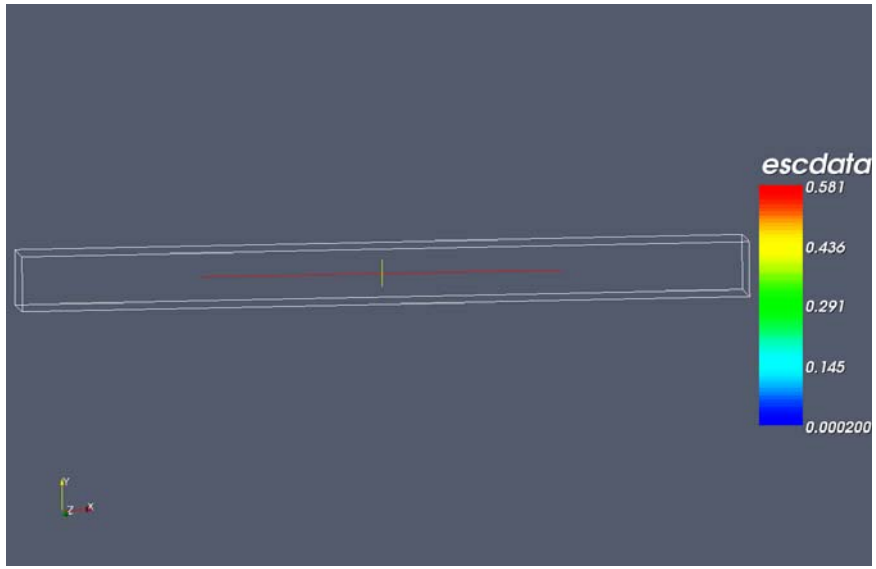
Results for the kinetic energy interpolation approach

- 11 Hydrogen atoms.
- There are a few domain boundary artifacts but do not exceed 2% of peak.
- Investigation in superior interpolation techniques is warranted .



Numerical Results: 3D Simulations

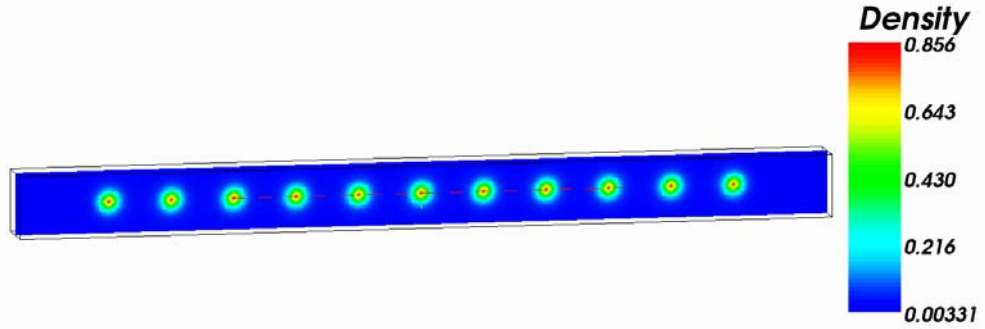
- Same Hydrogen string problem, but in 3D:
  - Parallel function/gradient evaluation
  - Parallel optimization solver
  - Constant mesh size
  - Dimension of problem: 35,672
  - Example run on Linux cluster, using 13 MPI processes
  - Note that both the cost per iteration and the number of iterations decreases with less active subdomains



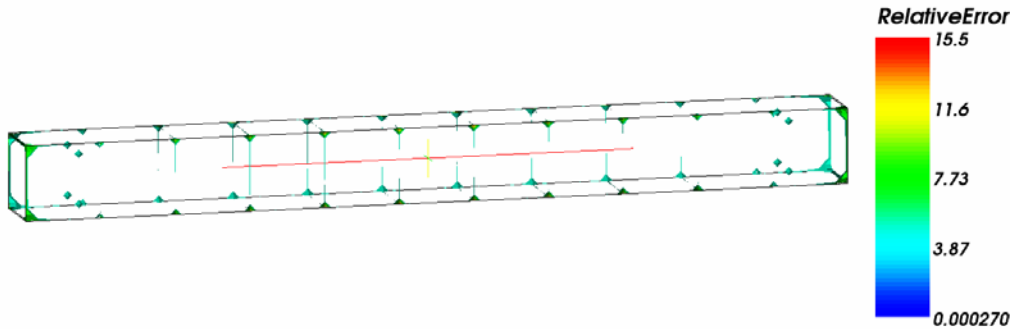
Active Subdomains	13	7	5
Number of Iterations	605	245	221
Total Energy	-14.257	-14.256	-14.256

Numerical Results: 3D Simulations

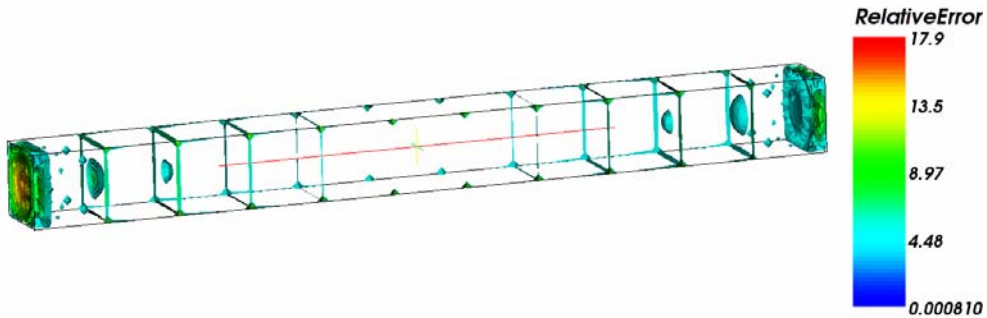
- 13 parallel processes
- 13 active subdomains



- 13 parallel processes
- 7 active subdomains



- 13 parallel processes
- 5 active subdomains



Example for potential description case

- Problem minimize the energy function of 101 atoms with pairwise Lennard-Jones potential V and representative dof x_1 and atom 61 fixed. The positions of nonrepresentative DOF are obtained by linear interpolation from positions of nearby representative DOF

$$E(x) = \sum_1^A \sum_{j>i}^A V(r_i - r_j),$$
$$x_1 = (r_1; r_2; r_3; r_4; r_{23}; r_{42}; r_{61}; r_{80}; r_{99}; r_{100}; r_{101})$$
$$g_1(x_1) = r_{61} - 61,\ g_2(x_2) = \emptyset,\ g_3(x_1, x_2) = \emptyset.$$

- Problem is solved with SNOPT in through AMPL, solution of (O) takes about 10 iterations.
- It can be verified from the outset that all assumptions (RECF), (ROCF) and (CSC) concerning the constraints are satisfied.
- At the solution it turns out that (SOSC) and the assumption that the interpolation ansatz is accurate are also satisfied; which means that the well posedness of the “interpolate and optimize” (RO) problem is ensured.
- But how about the HT constraint and “optimize and interpolate”?

Verification of the HT assumption

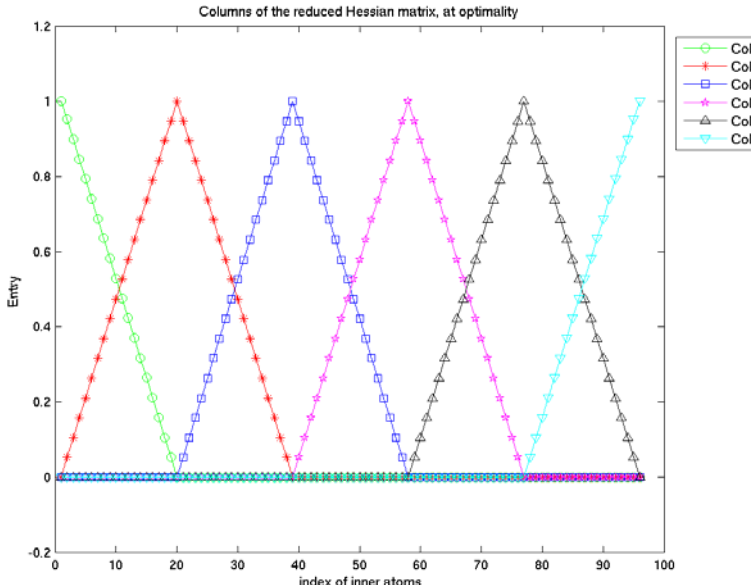
- Recall, the assumption stated that

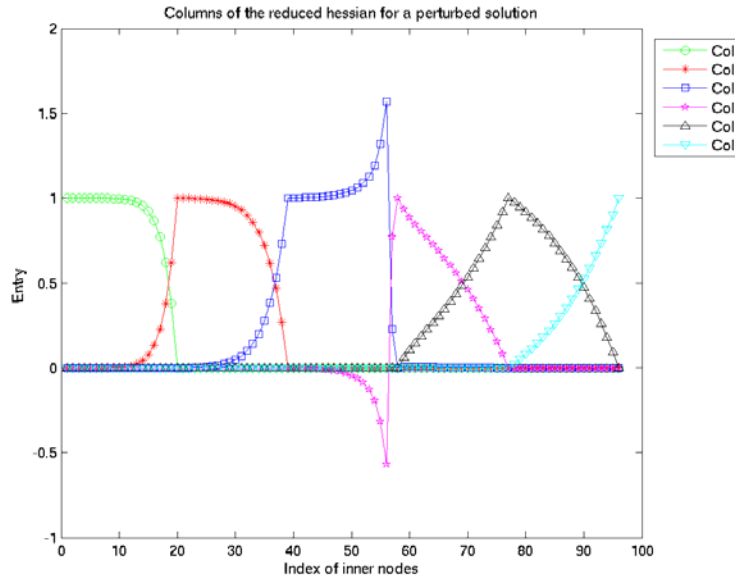
$$\left\| \nabla^2_{x_2 x_2} L(x^*, \lambda^*) T + \nabla^2_{x_2 x_1} L(x^*, \lambda^*) \right\| \leq \epsilon$$

- In top figure we plot the columns corresponding to DOF used in linear interpolation of

$$-\nabla^2_{x_2 x_2} L(x^*, \lambda^*)^{-1} \nabla^2_{x_2 x_1} L(x^*, \lambda^*)$$

- Note that the match with T is nearly perfect and our theory can be applied to ensure that (RE) is regular.
- But that does not follow solely from the atoms being positioned as a smooth function of the macroscale! See second figure where maximum relative interdistance perturbation is 1.6%





Example for electronic structure reconstruction.

- 1D example:
  - HT assumption is no longer verified at optimality, though convergence and stability of the “optimize and interpolate” case (RE) can be observed.
  - All assumptions for the “interpolate and optimize” case (RO) are satisfied.
- 3D example:
  - The (RE) approach was not coded.
  - The (RO) approach satisfies all assumptions except (SOSC) which we did not test, since we did not compute Hessians.
  - Note that the novelty here is also in the interpolation rule itself.

Conclusions and future work

- We have designed a nonlocal QC-like model reduction for DFT, and we have shown that it is accurate.
- We have given conditions for well posedness of the reduced problem, and show that they are reasonable for many configurations
- To do .. A lot
  - Test the approach for more realistic DFT approaches (OFDFT which includes gradients terms as well, Kohn Snam).
  - Better Interpolations which avoid artifacts at boundary,
  - Inequalities (though properties of reduced problem are reproduced faithfully if only rep DOF are constrained in original model).
  - Compress the long range interaction operators kernels using multipole or multiresolution, or discuss reduced Poisson Solves.
  - Determine weaker conditions of well-posedness for “optimize and interpolate”; force-based approaches.
  - Does the problem and choice of representative domains dictate the interpolation operator?