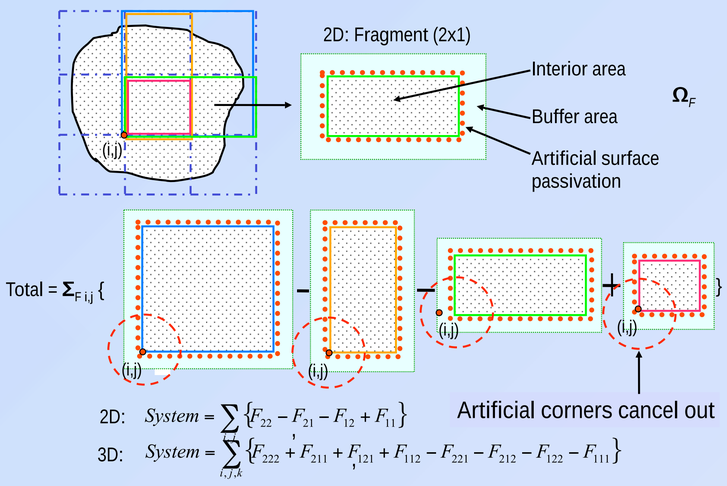
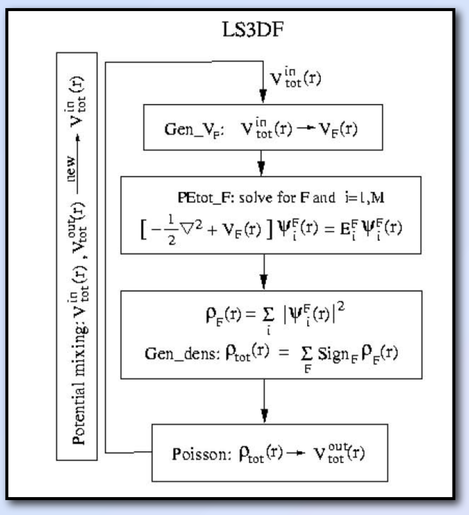
A New O(N) Method for Petascale Nanoscience Simulations

Zhengji Zhao, Juan Meza, and Lin-Wang Wang
Computational Research Division, Lawrence Berkeley National Laboratory

Nanostructures have been proposed for many applications including solar cells for renewable energy, biomedical imaging, and other novel materials. To fully explore these ideas, petascale *ab initio* materials simulations will be required. We have developed a new linear scaling 3 dimensional fragment (LS3DF) method, which is capable of simulating tens of thousands of atoms and is thousands of times faster than comparable direct local density approximation (LDA) calculations. LS3DF is based on a divide-and-conquer approach, which incorporates a novel patching scheme that effectively cancels out the artificial boundary effects due to the subdivision of the system. As a result, the LS3DF method scales up to thousands of processors and has excellent numerical agreement with direct LDA calculations. We have tested our method on different computer platforms, including the NERSC IBM SP3, Cray-XT4, and the CCNL Cray-XT3/XT4 machines. In addition, we have applied the LS3DF method to the computation of dipole moments of CdSe quantum dots providing new insights on a decades old problem in physics.





A comparison to a direct LDA calculation on a 339 atom silicon quantum dot and a 178-atom CdSe nano-rod

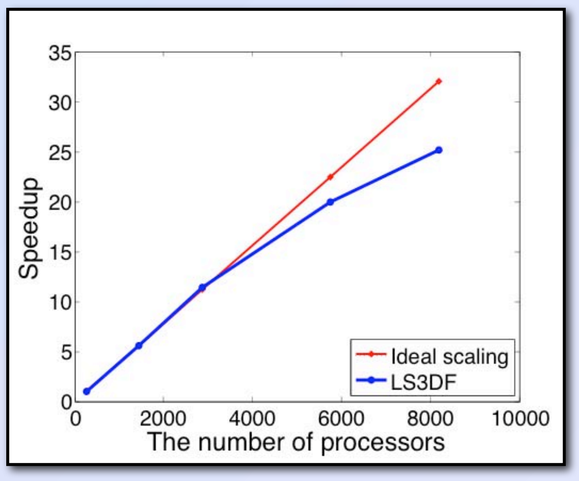
- Total energy difference: 0.1 kcal/mol (better than the typical 0.1eV error introduced by other numerical approximations: PW cut off, pseudopotential)
- Charge density difference: 0.2%
- Atomic force difference: 10^{-5} a.u (an order of magnitude smaller than the typical stopping criterion for atomic relaxation)
- The dipole moment difference: 1.3×10^{-3} Debye/atom.

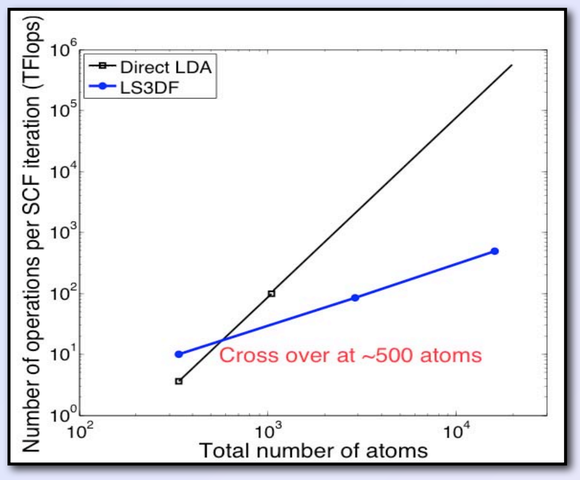
LDA calculations (Kohn-Sham equation) need to address quantum mechanical effects as well as classical electrostatic interactions. The LS3DF method is based on the observation that quantum mechanical effects are near-sighted, while classical electrostatic interactions are long range. Thus, we can solve for the quantum mechanical wavefunctions locally (we divide the system into fragments and solve each fragment independently), and then solve a Poisson equation globally, using the charge density computed by combining each of the fragment contributions. As shown in the flow chart of LS3DF above (right), the fragment Kohn-Sham equations (the second box in the flow chart) are solved independently for each fragment. This makes the computation perfectly parallelizable. The number of fragments is proportional to the size of the system, which means the method will scale linearly with the problem size.

The scheme for dividing a system into 3D fragments, and patching them together to get the total charge density (left) is a key element in LS3DF method. In 3D, at each (i,j,k) corner, there will be 8 fragments of different sizes (2x2x2, 2x2x1,2x1x1,1x1x1, etc.) shown as the dashed line boxes. The appropriate summation of the fragments using both positive and negative terms generates the cancellation of division boundary effects.

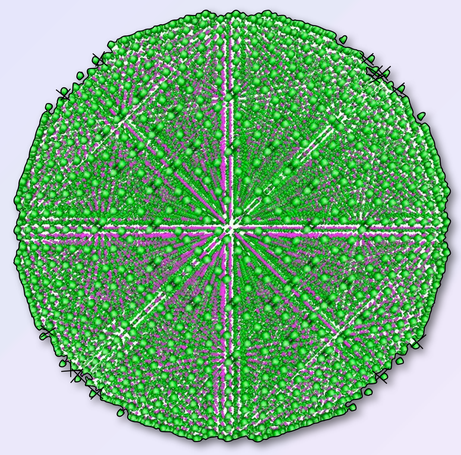
fragment size	0.5a	1a	1.5a
ΔE (meV/at)	30	2.9	4.0
$\sum_F \alpha_F \int \Delta V_F \rho_F dr$ (meV/at)	213	5.5	1.0
$\Delta\rho$	1.1%	0.14%	0.08%

LS3DF error convergence with respect to the fragment sizes. The fragment size 0.5a, 1.0a, and 1.5a corresponds to 8, 64, and 216 atoms in the 2x2x2 fragments. The third row in the table corresponds to boundary cancellation error, and the fourth row is the charge density error. The LS3DF error decays rapidly as the size of the fragment increases.

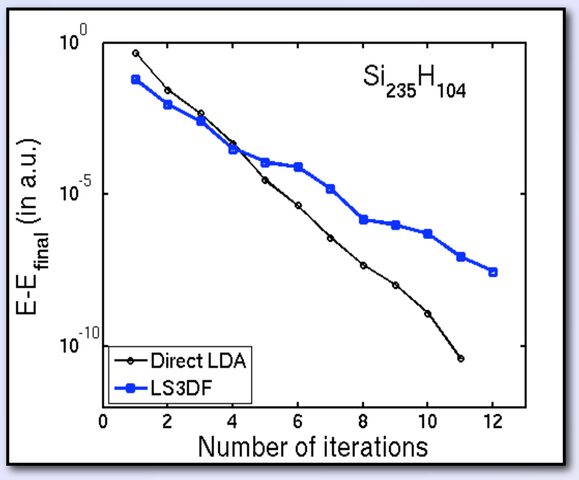


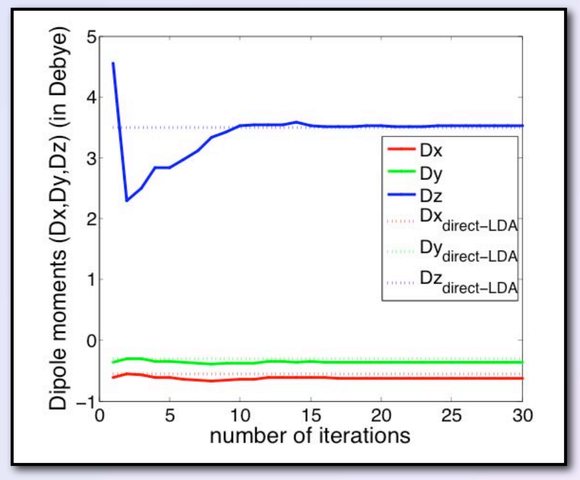


LS3DF scales almost linearly up to ~8000 processors on Cray XT4 (left, for a fixed size 2000-atom CdSe quantum dot). The cross over size between the LS3DF method and the cubic scaling direct LDA method is about 500 atoms, similar to other linear scaling methods.

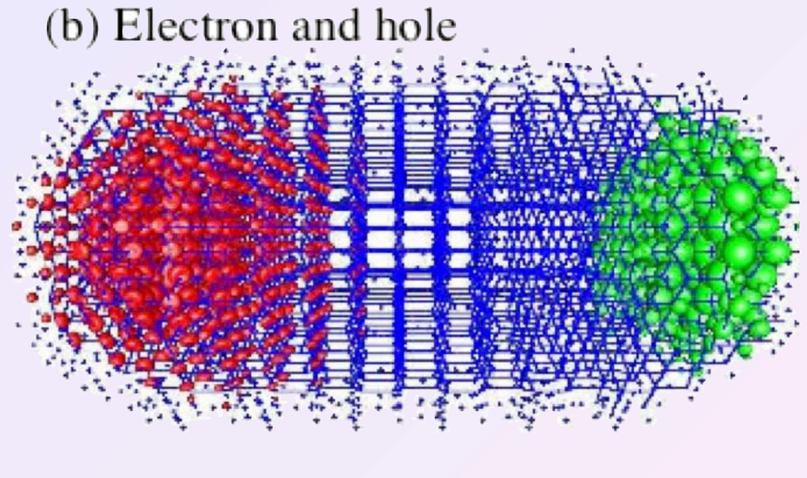


The charge density of a 15,000 atom quantum dot, $Si_{13607}H_{2236}$. Using 2048 processors at NERSC IBM SP3 machine, the calculation took about 5 hours, while a direct LDA calculation would have taken a few months.





The energy convergence (left) and dipole moment convergence (right) in the self-consistent iteration using the LS3DF method. The convergence rate (left) is similar to the direct LDA calculation and avoids the common convergence problem in other linear scaling methods. The LS3DF dipole moments (for a 178-atom CdSe quantum dot) convergent to the direct LDA values, demonstrating the accuracy of the LS3DF method.



The dipole moment in a CdSe nanorod produces an internal electric field, which localizes the electron (red) and hole (green) wavefunctions. The internal electric field (calculated here using the LS3DF method) plays a significant role in determining the electronic structure of nanosystems.

Funding provided by Department of Energy Advanced Scientific Computing Research and Basic Energy Sciences Nanoscience Initiative.
This research used resources of the National Energy Research Scientific Computing Center (NERSC), and National Center for Computational Sciences (NCCS), which are supported by the Office of Science of the U.S. Department of Energy