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# O(N) complexity algorithms for First-Principles Electronic Structure Calculations

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## Disclaimer



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# O(N) complexity algorithms for First-Principles Electronic Structure Calculations

The fundamental equation governing a non-relativistic quantum system of N particles is the time-dependant Schrödinger Equation [Schrödinger, 1926]

$$i\hbar \frac{\partial}{\partial t}\psi(x_1,...,x_N,t) = -\frac{\hbar^2}{2m}\Delta\psi(x_1,...,x_N,t) + V(x_1,...,x_N,t)\psi(x_1,...,x_N,t)$$

In 1965, Kohn and Sham proposed to replace this original many-body problem by an auxiliary independent-particles problem that can be solved more easily (Density Functional Theory). Solving this simplified problem requires to find the subspace of dimension N spanned by the N eigenfunctions $\psi_i$ corresponding to the N lowest eigenvalues $\varepsilon_i$ of a non-linear Hamiltonian operator $\hat{H}$ determined from first-principles

$$\hat{H}\psi_i(r) = \varepsilon_i\psi_i(r)$$

From the solution of the Kohn-Sham equations, forces acting on atoms can be derived to optimize geometries and simulate finite temperature phenomenon by molecular dynamics. This technique is used at LLNL to determine the Equation of State of various materials, and to study biomolecules and nanomaterials.

## *Scaling problem:*

- Standard computational algorithms represent the electrons by N quantum wave functions extended over the whole computational domain (see Fig. 1)
- This leads to $O(N^2)$ storage requirements and $O(N^3)$ arithmetic operations
- Using LLNL powerful super-computers, large problems can be solved. But this cubic scaling becomes a critical bottleneck which limits our capabilities to study larger physical systems

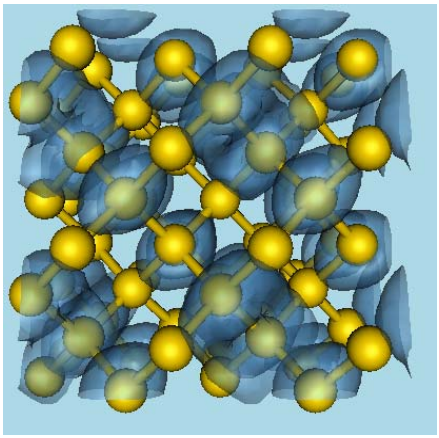

**Figure 1: Isosurface of a computed electronic wave function in silicon crystal (64 atoms cell). It limits the volume in which one has the highest probability of finding the electron.**

## *Maximally localized Wannier functions representation*

- The electronic structure can be efficiently represented by a set of "localized" orbitals with a limited spread independent of the system size ("Maximally Localized Wannier Functions")
- These functions can be obtained from the eigenfunctions of the Hamiltonian operator by an orthogonal transformation (see Fig. 2)

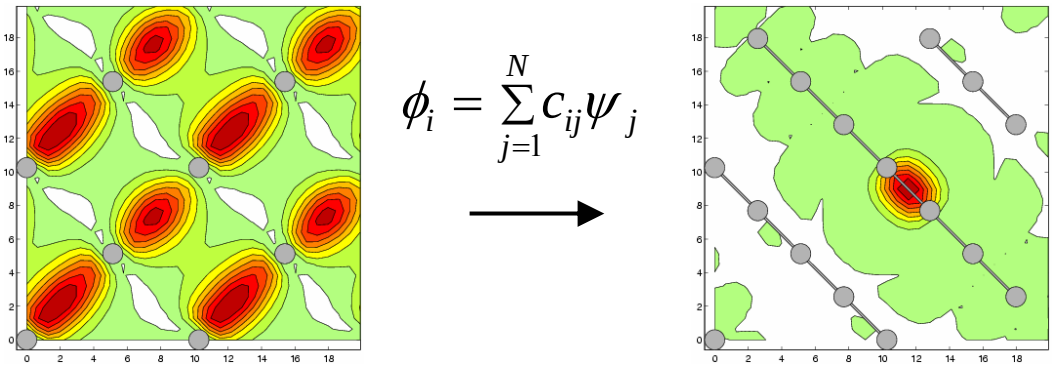


**Figure 2: Electronic orbital in silicon bulk (contour plot in slicing plane with projections of nearest atoms). A linear combination of eigenfunctions can lead to very localized functions.**

## *O(N) algorithm:*

- Represent electronic orbitals on real-space uniform mesh and use finite differences discretization
- compute directly localized orbitals (truncated beyond a cutoff radius) by minimizing energy functional with localization constraints that only marginally affect accuracy
- This approach is justified by the theory of "Maximally Localized Wannier Functions"

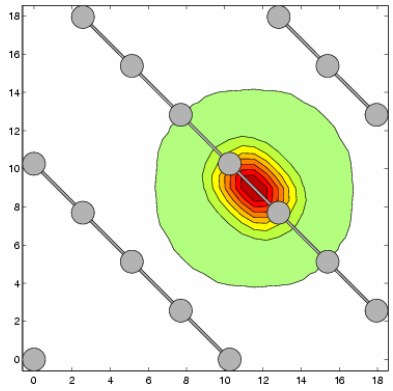


**Figure 3: strictly localized orbital in silicon crystal directly optimized on real-space mesh with localization constraints (contour plot in slicing plane)**

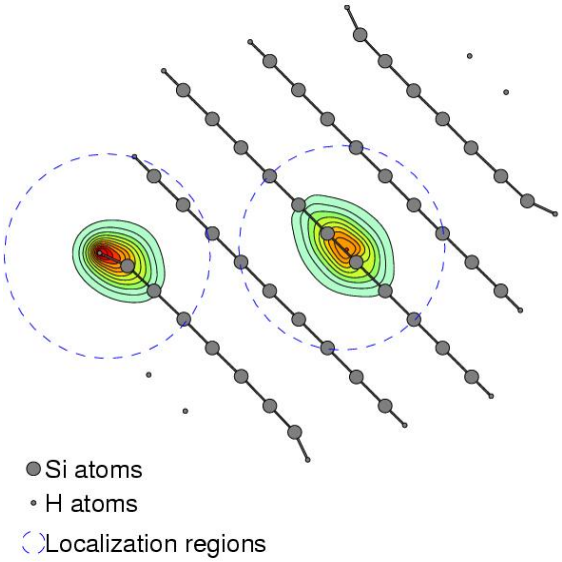


**Figure 4: 2 localized orbitals in silicon nanowire (slice perpendicular to wire axis)**

## *Overcoming the cubic scaling wall*

We have demonstrated that this “localized orbitals” technique becomes more efficient than the traditional Plane Waves approach for physical systems larger than ~500 atoms.

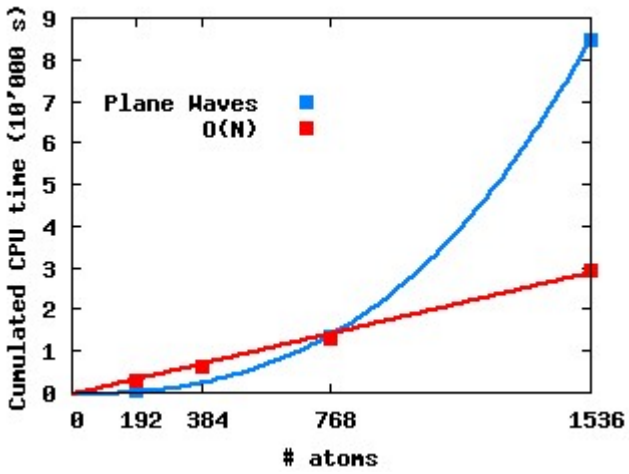


**Figure 5: comparing computer time for the standard PW approach (QBox code) and our new linear scaling algorithm for a quantum simulation of liquid water (from Ref. [5])**

## *O(N) Finite Difference Code: MGmol*

- Author: J.-L. Fattebert
- ~50K lines C++
- Parallel: scales on more than 1000 CPUs
- Based on domain decomposition with nonoverlaping localized orbitals treated in parallel
- Libraries:
  - MPI
  - PB (own multigrid library for Poisson solver and preconditioner)
  - ScaLAPACK (with C++ interface)
  - HDF5 (parallel)

## *Finite element approach for calculations on locally-refined meshes (with Rich Hornung http://www.llnl.gov/comp/bio.php/hornung1)*

As an alternative to Finite Differences, one can use a Finite Element approach as a real-space discretization for the Kohn-Sham equations. In our experience, FE is somewhat more expensive than FD, but its variational principle is convenient when working with non-uniform meshes.

We have developed a new electronic structure code based on an existing AMR parallel software infrastructure, SAMRAI (Structured Adaptive Mesh Refinement Application Infrastructure, http://www.llnl.gov/casc/SAMRAI/). SAMRAI is a C++ object-oriented AMR support infrastructure developed at LLNL. It provides flexible software tools for a wide range of AMR application research: parallel data infrastructure, gridding algorithms and communications functionalities.

We use a hierarchical quadratic Finite Elements approach [6].

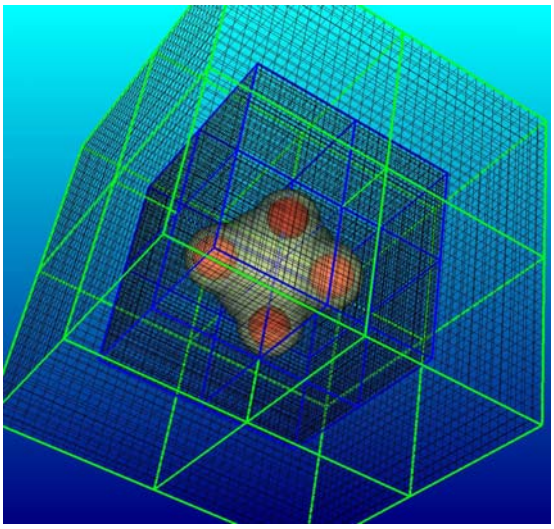

**Figure 6: Illustration: electronic density isosurface for Be4 cluster, domain decomposition for 8 CPUs**

## *Using Adaptive Mesh Refinement to improve scaling while controlling accuracy*

Maximally Locallized Wannier functions representation can benefit from local mesh refinement by representing their tails on a mesh coarser than the one used for their centers (Fig. 7). To investigate this new idea, new functionalities have been implemented in SAMRAI to allow a different patch hierarchy for each orbital. In practice a single patch hierarchy is used, but functions are set "inactive" on some patches (no data allocated and no work done). This is the concept of "Locally-Active Data" (Fig. 8). An electronic structure code is being developed based on this idea.

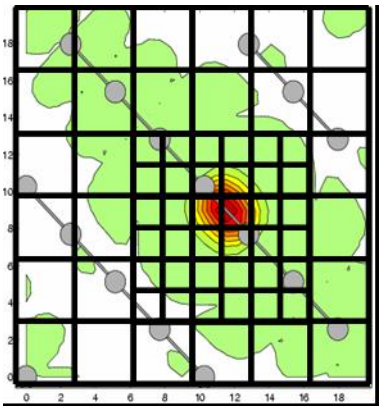


**Figure 7: Maximally Localized Wannier Function on a locally-refined grid**

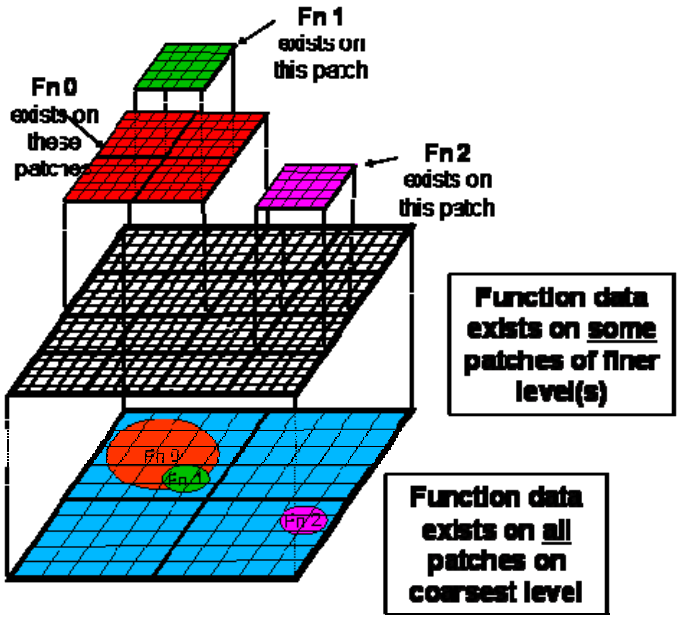


**Figure 8: concept of "Locally-Active Data": single patch hierarchy, but functions "inactive" on some patches (no data allocated).**

## *Links:*

SciDAC project: Quantum Simulations of Materials and Nanostructures
http://angstrom.ucdavis.edu/scidac/

SAMRAI
http://www.llnl.gov/casc/SAMRAI/

## *References:*

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[4] J.-L. Fattebert and F. Gygi, *Linear scaling first-principles molecular dynamics with controlled accuracy,* Comput. Phys. Comm., 162, no 1 (2004), pp. 24-36.

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[6] J.-L. Fattebert, R.D. Hornung and A.M. Wissink, *Finite element approach for density functional theory calculations on locally-refined meshes*, J. Comput. Phys., in Press.

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