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LS3DF for Large-Scale Electronic Structure Calculations (2008)

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The new Linear Scaling Three-Dimensional Fragment (LS3DF) method is hundreds to thousands of times faster than direct density functional theory calculations for large systems (10,000–100,000 atoms), yet yields essentially the same numerical results. LS3DF has achieved a speed of 107.5 Tflop/s on 131,072 cores of an IBM BlueGene/P system, making it the first linear scaling* ab initio *electronic structure code that has been efficiently parallelized to such a large number of processors. In recognition of this achievement, the LS3DF developers have been named finalists in the ACM Gordon Bell Prize competition.

There are many material science and nanoscience problems involving thousands to tens of thousands of atoms that can be accurately simulated only by *ab initio* self-consistent methods. These include nanostructures such as quantum dots and wires, core/shell nanostructures, as well as more conventional systems such as semiconductor alloys.

For example, materials that have separate electron states within the energy band gap (mid-band-gap states) have been proposed as second-generation solar cells. Such systems could theoretically increase solar cell efficiencies from 40% to 63%. One potential way to produce mid-band-gap state materials is to use substitutional semiconductor alloys such as zinc tellurite oxide (ZnTeO). But the actual characteristics of these mid-band-gap states are still unknown.

The properties of mid-band-gap states can be determined by *ab initio* density functional theory (DFT) calculations. However, due to the small percentage of oxygen atoms, large supercells containing thousands of atoms must be used to describe the random distribution of the oxygen atoms properly. This makes calculations using a direct DFT method impractical; for example, a 13,824-atom ZnTeO supercell would require four to six weeks or more of run time on 20,000 processor cores.

In order to solve such problems and to take full advantage of future computer systems now on the drawing boards that will employ hundreds of thousands of cores, Lin-Wang Wang and colleagues in the Computational Research Division at Lawrence Berkeley National Laboratory have developed a new linear scaling method that also exhibits excellent parallel scalability. For the 13,824-atom system mentioned above, this method, called LS3DF for Linear Scaling Three-Dimensional Fragment, is roughly 400 times faster than conventional $O(N^3)$ methods (e.g., QBox, Paratec, PEtot, and VASP). This new scheme produces a fully converged result within 2–3 hours, yet yields essentially the same numerical results as the conventional $O(N^3)$ methods. Since it uses a divide-and-conquer scheme, this method is well suited for large-scale computations.

The LS3DF algorithm scaled almost perfectly to 17,280 cores of the NERSC dual-core Cray XT4 (Franklin), and a performance analysis indicated that LS3DF would scale to a much larger number of cores without any substantive algorithmic obstacles. On 17,280 cores, LS3DF achieved 32 Tflop/s or 32% of the peak floating-point performance of the machine. In a separate test, LS3DF ran on 30,720 cores on the quad-core Cray XT4 (Jaguar) at the Oak Ridge Leadership Computing Facility, reaching 60 Tflop/s at 23% of the theoretical peak. In a later run on the IBM BlueGene/P system (Intrepid) at the Argonne Leadership Computing Facility, the code achieved 107.5 Tflop/s on 131,072 cores, or 24.2% of peak. This makes LS3DF the first variationally accurate linearly scaling *ab initio* electronic structure code that has been efficiently parallelized to such a large number of processors.

In recognition of this achievement, the LS3DF developers have been named finalists in the ACM Gordon Bell Prize competition. The winner will be announced on Thursday, November 20, 2008 at the SC08 Conference.

The LS3DF method uses a novel decomposition and patching scheme to do *ab initio* DFT calculations for large systems. This method cancels out the artificial boundary effects that arise from the spatial decomposition. As a result, the LS3DF results are essentially the same as the original full-system DFT results, with errors smaller than those introduced by other sources of numerical approximations. The fragments of the LS3DF algorithm can be calculated separately with different groups of processors, which leads to almost perfect parallelization on tens of thousands of processors. The resulting computational times are thousands of times smaller than conventional DFT methods, making calculations with 100,000 atom systems possible.

A divide-and-conquer scheme is a natural approach for mapping the physical locality of a large problem to the architectural locality of a massively parallel computer. LS3DF is based on the observation that the total energy of a given system can be broken down into two parts: the electrostatic energy and the quantum mechanical energy (i.e., kinetic and exchange-correlation energy). While the electrostatic energy is long-range and must be solved via a global Poisson equation, the computationally expensive quantum mechanical energy is short-range and can be solved locally. The idea is to divide the whole system into small fragments, calculate the quantum mechanical energies of these fragments, and then combine the separate fragment energies to obtain the energy of the whole system.

A critical issue in a divide-and-conquer scheme such as this is how to combine (patch) the fragments. The core of LS3DF is a novel patching scheme that cancels out the artificial boundary effects caused by the division of the system into smaller fragments. As a result of this cancellation, the results are essentially the same as a direct calculation on the large system. In LS3DF, once the fragment sizes are chosen to obtain a given numerical accuracy, the computational cost is proportional to the number of fragments—hence the name, Linearly Scaling Three-Dimensional Fragment method. In contrast, the cost of conventional methods scales as $O(N^3)$, where N is the size of the system in atoms.

Using a small group of cores to solve the quantum mechanical part of each fragment independently, LS3DF also scales in performance almost perfectly with the number of cores. Only a small overhead is needed to patch the fragment charge densities into a global charge density, and to solve the Poisson equation for the whole system. As a result, this method can be employed on computer systems with tens of thousands of cores.

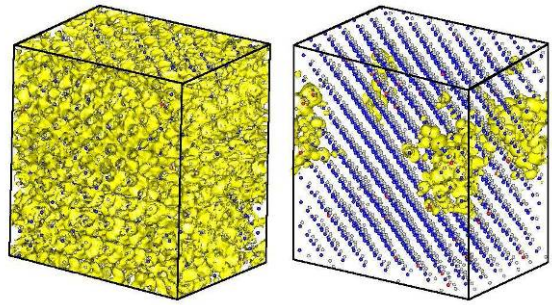

Fig. 1. Isosurface plots (yellow) of the electron wavefunction squares for the bottom of the conduction band (left) and the top of the oxygen-induced band (right). The small grey dots are Zn atoms; the blue dots are Te atoms; and the red dots are oxygen atoms.

The simulation of a 13,824-atom ZnTeO supercell yielded substantive scientific results. First, the researchers found that there is a 0.2 eV band gap between the ZnTe conduction band and the oxygen-induced band, implying that this alloy can be used for solar cell application. Secondly, they found that the oxygen-induced states form a very broad band (0.7 eV) inside the band gap of ZnTe. As a result, the simulations predict that the theoretical maximum efficiency of solar cells made from this alloy will most likely be lower than the 63% figure estimated based on a narrow mid-band-gap. Also, as shown in Fig. 1, the oxygen-induced states can cluster among a few oxygen atoms. Such clustering will have an impact on the mobility of the electrons in those states.

The LS3DF method is applicable to insulator and semiconductor systems, which covers a current gap in the Department of Energy's materials science code portfolio for large-scale *ab initio* simulations. The performance results demonstrate the potential for petascale computation using the LS3DF method.

Publications:

L. W. Wang, B. Lee, H. Shan, Z. Zhao, J. Meza, E. Strohmaier, and D. Bailey, "Linear scaling 3D fragment method for large scale electronic structure calculations," LBNL technical report LBNL-603E; Gordon Bell paper, SC08. http://repositories.cdlib.org/lbnl/LBNL-603E/

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