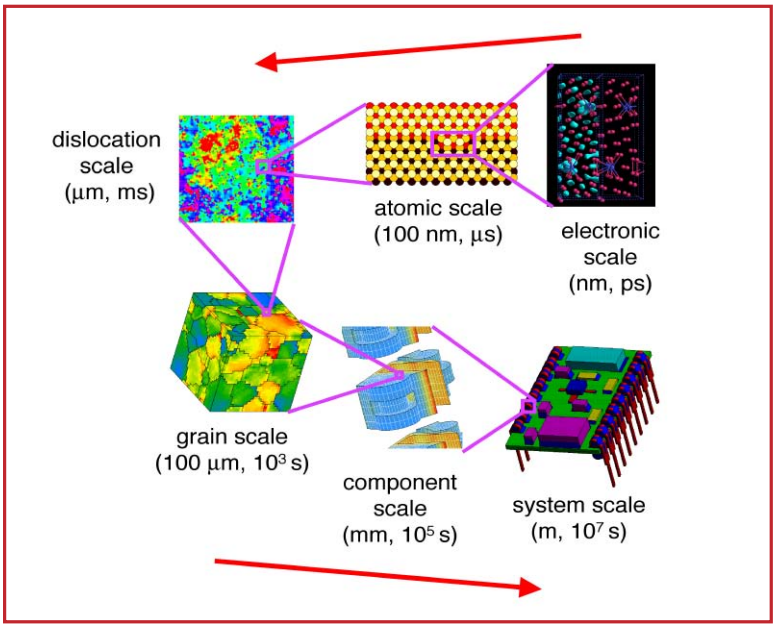
Computers and Information Sciences

Multiscale Computational Material Methods



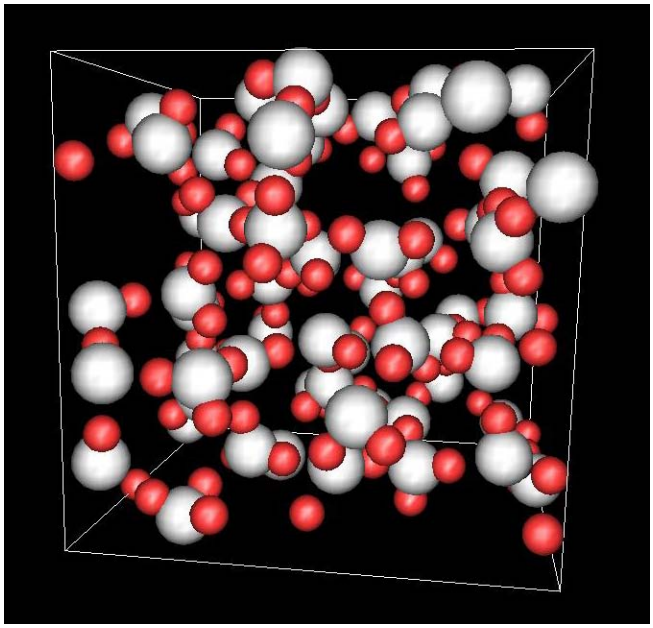


Figure 1: (left) DFT calculations on the electronic scale are the underpinning of many predictive multi-scale efforts at Sandia. (Image courtesy E. Holm).
Figure 2: (right) To obtain a correct description of water, not only are large supercells needed, but also molecular dynamics simulations over tens of pico seconds are needed in order to account for temperature effects. This requires an XC functional that is both fast and accurate. The picture shows a snapshot of a DFT-MD simulation using AM05 for 64 water molecules.

The AM05 density functional enables accurate and fast quantum simulations of condensed matter systems.

Enabling Predictive Multiscale Modeling and Simulations

Quantitative results for a broad range of systems in combination with a relatively low computational cost have made density functional theory (DFT) the foundation of most large-scale quantum mechanical simulations in science. Thus DFT is the workhorse method for high-fidelity quantum calculations at the electronic scale that are the basis for many predictive material simulations at larger scales (Figure 1). DFT simulations are helping solve some of the most difficult materials problems: describing radiation effects in semiconductor materials; materials behavior under extreme conditions, such as that encountered in high energy-density physics experiments at Sandia's Z-accelerator; or unveiling how ion selection occurs in biological systems.

At the core of each DFT electronic energy calculation lays the exchange-correlation (XC) functional, which sets the limit for the accuracy of the calculations. Theory tells us that a "divine" XC functional exists with which DFT calculations provide the correct electronic ground state. Because this functional is not known, many approximate functionals must have been developed. The lack of systematic improvement in functionals for solid-state systems has made it difficult to predict which one is the most accurate for a given problem. The development of new functionals is thus critical to the progress of not only computational materials science, but also that of physics and chemistry in a broad sense. Since DFT is increasingly being employed for large systems (Figure 2) of several hundred atoms, and for long molecular dynamics simulations (tens of ps), the tradeoff between speed and

Technical Contact:
Ann Mattsson, Pd.D.
505-844-9218
aematts@sandia.gov

Science Matters Contact:
Alan Burns, Ph.D.
505-844-9642
aburns@sandia.gov

Sandia National Laboratories

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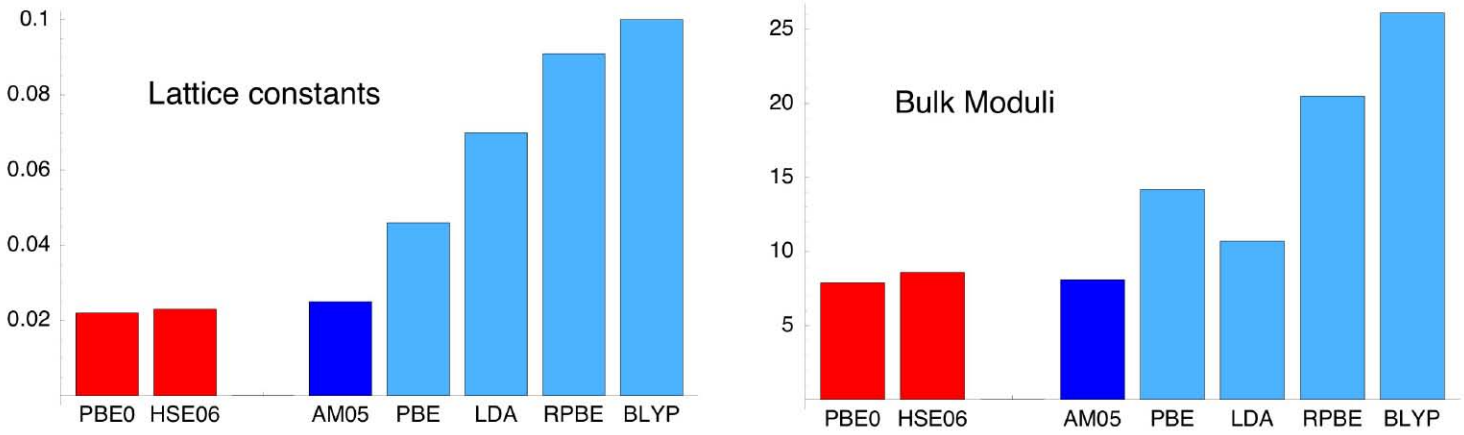

Figure 3: Comparison of mean absolute errors (MAE) for properties of 20 solids calculated with seven different functionals. The left panel shows lattice constants MAE in Ångström (0.1 nm) and the right panel shows bulk moduli MAE in GPa. Generalized gradient approximation type functionals (blue) are one to three orders of magnitudes faster to use than hybrids (red). AM05 has the same accuracy as hybrids for solids, and thus enable accurate and fast DFT calculations of defects in semi-conductors. It also allows for the use of DFT-MD as an accurate tool in Equation of State construction.

accuracy is arising as an additional major concern in functional development.

There is a fundamental difference between the behavior of electronic wave functions in a bulk solid and regions at surfaces and in interstitial regions in semiconductors. In the former, the wave functions are periodic propagating waves while in the latter, they are damped evanescent surface waves. By first identifying, and then focusing on this duality, we succeeded in developing a functional with high accuracy while retaining low complexity and thus high speed.
For solids, the AM05 (Armiento and Mattsson, 2005) XC functional is proving to be as accurate as the best available XC functionals (Figure 3), while allowing one to three orders of magnitudes faster calculations. A calculation of a defect in a semiconductor like GaAs that runs overnight (12 hours) using AM05 would take almost six weeks using the fastest of the accurate functionals used up to now (HSE03/HSE06). Hence, AM05 is emerging as the functional of choice for accuracy as well as speed for essentially all materials.

Even so, there is more work to be done. Neither AM05 nor any other existing functional can treat dispersive forces or van der Waals binding, such as are prevalent in two important classes of materials, biomolecules and molecular solids, the latter of which includes high explosives and energetic materials. At Sandia, there is a focus on rational compound design, where it is of utmost importance to be able to obtain the right trends so that a target property can be calculated as a function of chemical composition. These applications require yet another leap in understanding and efficiency.

Publications:

"The AM05 density functional applied to solids", Ann E. Mattsson, Rickard Armiento, Joachim Paier, Georg Kresse, John M. Wills, and Thomas R. Mattsson, *Journal of Chemical Physics*, to be published 07 February 2008.

"Functional designed to include surface effects in self-consistent density functional theory", Rickard Armiento and Ann E. Mattsson, *Physical Review B* **72**, 085108 (2005).

"In Pursuit of the 'Divine Functional' ", Ann E. Mattsson, *Science* **298**, 759 (25 October 2002).

"Designing meaningful density functional theory calculations in materials science—a primer," Ann E. Mattsson, Peter A. Schultz, Michael P. Desjarlais, Thomas R. Mattsson, and Kevin Leung, *Modelling and Simulation in Materials Science and Engineering* **13**, R1-R31 (2005).

Sandia National Laboratories

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