MAR10-2009-002591

Abstract for an Invited Paper
for the MAR10 Meeting of
the American Physical Society

Advances in Orbital-Free Density Functional Theory: Physics and Algorithms[1]

EMILY CARTER, Princeton University

Orbital-free density functional theory (OFDFT) is a first principles quantum mechanics method that can scale linearly with system size by solving directly for the electron density instead of introducing an auxiliary set of one-electron orbitals as is done in conventional Kohn-Sham (KS) DFT. Orbitals must be kept orthonormal; imposing this constraint involves a cubically scaling step. KSDFT can be made to scale linearly beyond a crossover point within a localized orbital framework and hence molecules and insulators can be made to scale linearly within KSDFT. Metals generally do not exhibit linear scaling within KSDFT due to their inherently delocalized electronic structure. OFDFT offers an alternative scheme by introducing a kinetic energy density functional (KEDF) and local electron-ion pseudopotentials (LPSs). Thus the accuracy of OFDFT depends on the representation of these two terms. We now have a routine tool for constructing accurate LPSs by inverting the KS equations for bulk crystals. These BLPSs are validated against accurate nonlocal PSs within KSDFT. A decade ago, we reported a nonlocal KEDF that accurately captures the physics of nearly-free-electron-like metals. Here we report a new nonlocal KEDF that explicitly contains the physics required to describe semiconductors. Extensive tests on silicon and a variety of compound semiconductors reveal this new KEDF to be accurate for many properties, thus opening up the door to accurate OFDFT calculations on semiconductors. We also report the systematic elimination of bottlenecks within our OFDFT code that render the entire algorithm linear scaling for all system sizes (no crossover point). With parallelization then introduced via domain decomposition, quantum mechanical simulations of metal samples containing up to 1 million atoms have been demonstrated using a modest number of processors. With this new capability we are studying mesoscale features that control mechanical properties of Al and Mg alloys.

[1]National Science Foundation