Electronic Structure of Semiconductors and Molecular Magnets

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Summary of Research:

Frank Petruzielo: Molecular calculations in quantum Monte Carlo frequently employ a mixed basis consisting of contracted and primitive Gaussian functions. While standard basis sets of varying size and accuracy are available in the literature, we demonstrate that reoptimizing the primitive function exponents within quantum Monte Carlo yields more compact basis sets for a given accuracy. Particularly large gains are achieved for highly excited states.

For calculations requiring non-diverging pseudopotentials, we introduce Gauss-Slater basis functions that behave as Gaussians at short distances and Slaters at long distances. These basis functions further improve the energy and fluctuations of the local energy for a given basis size. Gains achieved by exponent optimization and Gauss-Slater basis use are exemplified by calculations for the ground state of carbon, the lowest lying excited states of carbon with 5S0, 3P0, 1D0, 3F0 symmetries, carbon dimer, and naphthalene. Basis size reduction enables quantum Monte Carlo treatment of larger molecules at high accuracy [1].

Michael Lowe: Our research on Nanolab consists of three main projects. The first two consist of predicting properties of species involved in lithium-sulfur batteries, and the third project was a quick structural analysis of a novel nitride. Lithium-sulfur batteries have the highest theoretical energy density of any secondary battery system, but implementation is limited due to a complex discharge mechanism. We are using Gaussian03 to predict reduction potentials for a range of polysulfide species, to complement ongoing electrochemical and spectroscopic data about key polysulfides involved in battery operation. Although there are significant differences between the computational and electrochemical environment, we have demonstrated that in the cases with experimental data, the DFT-predicted reduction potentials correlate with the observed values. We are also using a finite-difference method approach to predicting spectroscopic transitions (FDMNES) to predict the x-ray absorption signal near the sulfur K-edge for select sulfur-based compounds. This modeling has enabled us to resolve some unexpected features in the x-ray absorption spectroscopy of lithium sulfide.

The third project uses DFT within the code VASP to predict the ground-state structure of some nitride/oxide compounds. The recently-synthesized AgTaN2 is the first crystal to report covalent Ag-N bonding, and the computed structure for the nitride was used to demonstrate that the reported lattice parameters and positions are reasonable and thermodynamically stable. The results have been included in a paper that will be submitted for publication soon.

References: