Title of Project: *Scalable Computational Tools for Discovery and Design -- Excited State Phenomena in Energy Materials*

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Scope of work: The objective of this project is to develop and implement new methods and theories to elucidate and predict excited electronic state phenomena in energy related materials, *e.g*., materials for photovoltaic and photocatalysis. Understanding excited state phenomena requires knowledge of both the ground state properties and the related many-electron interactions with excited states, which may involve structural and orbital relaxations. As such, quantitative predictions for excited state phenomena are at the leading edge of current theories for the electronic structure of materials. Our approach will be based on advanced algorithms and many-body perturbation theory to facilitate the *ab initio* calculations and prediction of quasiparticle excitations and lifetimes, optical spectra, excited-state energy surfaces, transport properties, and other excited-state properties/processes. We will employ a unified approach that will describe simultaneously complex ground state structures and excited state phenomena.

The methodology and algorithm development efforts will be based on two of the most advanced and accurate approaches in materials research. Excited-state properties will be addressed in an interacting Greens functions formalism within many-body perturbation theory. Quasiparticle spectra will be computed within the GW approximation for the electron self-energy, and two-particle optical excitations will be calculated within a Bethe-Salpeter equation approach, including electron-hole interactions. Ground-state properties will be addressed with pseudopotentials within density functional theory. Advances in both excited and ground state codes will be required to handle the wide range of materials problems associated with energy related applications.

Science applications will include molecular organic assemblies and transition-metal oxides, which are examples of two important classes of materials that promise low-cost, sustainable solar energy conversion. Although structurally distinct, these materials classes share common chemical attributes –highly-localized, sometimes strongly-correlated electronic states and, in some instances, appreciable noncovalent interactions. Additionally, interfaces (both metal-semiconductor and organic-organic) are crucial to energy conversion, and in many cases, their impact on device function is not well understood.

We will carry out studies on the following systems:
• Chromophore or dye molecules in gas-phase, in solution, or at a surface;
• Solvents, *e.g.* water or electrolyte, in contact with an oxide or metal electrode;
• Molecular junctions and self-assembled monolayers;
• Multicomponent inorganic semiconductor nanostructures;
• Transition metal oxides, with defects, dopants, and with magnetic cations;
• Organic molecular crystals and assemblies, and donor-acceptor molecular interfaces.

The research team includes physical scientists, materials engineers, computer scientists, and applied mathematicians at universities, government labs, and in SciDAC institutes. The research program will address computational challenges associated with energy materials in a comprehensive manner, drawing on the team's diverse set of skills, and will deliver scalable, open-source codes for ground state and excited state properties. These codes are targeted to be used with versatility and confidence across the scientific community and to handle complex problems associated with large numbers of atoms including magnetic species, defects, and disparate boundary conditions, *i.e.*, the central problems encountered in materials of relevance for the next-generation energy conversion and storage technology.