The U.S. Department of Energy SciDAC Partnership

Predictive Computing for Condensed Matter

University of Illinois at Urbana-Champaign
The College of William & Mary
and Princeton University

In collaboration with
University of Oregon and The SciDAC SUPER Institute

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Fundamental progress in energy technology depends critically on our ability to fabricate, characterize, and optimize advanced materials that can convert, transport, and store energies. To achieve this, we must be able to understand materials' properties from the first principles and to design new materials with predicted useful properties by harnessing the nonabating increase in computing power such as at Department of Energy's leadership supercomputers. It has been clear, however, that today's standard computational method for solids, i.e., density-functional theory, is fundamentally limited in its ability to predict response or excited-state properties as well as any properties of strongly correlated solids, which are often responsible for remarkable properties of advanced materials ranging from photovoltaic cells, light emitting diodes, electrodes in fuel cells, (photo)catalysts, to high-temperature superconductors. This may be contrasted to molecular sciences, where several hierarchies of systematic methods exist such as many-body perturbation and coupled-cluster methods. Efficient, robust, parallel software implementing these methods are widely available, enabling chemists to routinely predict virtually any observable at arbitrary high accuracy, transforming increasing wider areas of chemistry into a computational science.

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The same transformation is urgently needed for materials and chemical processes in condensed phases. The overarching goal of this SciDAC Partnership is, therefore, to perform a tightly integrated, collaborative research that aims at bringing this transformation. The team consists of researchers in the areas of theoretical chemistry, theoretical physics, and computer science as well as experimental condensed matter physics and conducts research in three key areas: theories, software, and applications:

Theories: We will establish novel electronic structure theories for solids (and liquids) that are *predictive* in the sense that the fidelity of simulation can be increased systematically––at increased computational cost—and hence the calculated results are guaranteed to converge at the exact solutions of the Schrödinger equation, the fundamental equation of motion of chemistry and materials; that can handle *excited electronic states* and thus response properties, going beyond the usual one-electron pictures; that can be applied to *strongly correlated* electronic structures by exposing hidden mathematical structures and symmetries therein. We will extend diagrammatic many-body methods such as many-body perturbation and coupled-cluster methods to solids in the ground and excited states and fully develop quantum Monte Carlo (QMC) methods for normal and strongly correlated solids, while exploring emerging mathematical descriptions of strongly correlated solids such as the density matrix renormalization group and its two- and three-dimensional extension known as tensor networks, dynamical mean-field theory, the use of QMC in determinant spaces such as auxiliary-field QMC and full configuration-interaction QMC, as well as the construction of *quantitative* model Hamiltonians by down-folding them.

Software: We plan to create a suite of software implementing the predictive electronic structure theories for solids (and liquids), which are, by virtue of exploiting *novel software technologies* and *new algorithms*, to be simultaneously *scalable*, *extensible*, and *maintainable* despite complex, diverse, and rapidly evolving supercomputer architectures. To achieve this, not only do we completely redesign algorithms of diagrammatic methods utilizing, for instance, the Monte Carlo integrations, which are more easily subjected to multi-level, fault-tolerant parallelism at petascale, we will also develop and deploy an innovative programming environment that allows us to write quality software in the shortest time that is scalable on a diversity of supercomputing platforms including mixed CPU-GPU systems. The programming environment consists in automating data layout marshaling, granularity adjustment, and data buffering for different types of processors, shielding programmers from architecture details. At the core of these automation capabilities lies our ability to measure the cost and performance of any program on a diversity of architectures, which will be furnished by the TAU toolsuite developed by the SciDAC SUPER Institute with which we partner.

Applications: We aim at applying these predictive methods as implemented in highly scalable parallel software to some of the *outstanding condensed matter problems*, including unexplained experimental results for high temperature superconductors, the excited states in graphene, etc., for which new experimental data obtained by the experimental solid-state physics investigator are available to the team. Comparison of computational and experimental results also serves as *mutual verification and experimental validation*. The successful outcome in any of these problems can fundamentally alter our understanding of condensed matter physics and may bring a scientific discovery through advanced computing.