QMCBD: A LIVING DATABASE TO ACCELERATE WORLDWIDE DEVELOPMENT AND USE OF QUANTUM MONTE CARLO METHODS

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EXECUTIVE SUMMARY

Blue Waters has enabled us to carry out automated, high-throughput quantum Monte Carlo calculations of condensed matter systems, and to create a database titled QMCDB (Quantum Monte Carlo DataBase) to share these results. We have been able to simulate approximately fifteen different classical semiconductors, which represents the largest set of QMC condensed matter simulations carried out to date. The database contains formation enthalpies and optical excitation energies, calculated via the highly accurate quantum Monte Carlo technique. QMCDB will serve as a platform for easy, searchable data exchange to accelerate the knowledge base around the use of QMC for materials modeling. This work would not be possible without Blue Waters, which allows the calculation of a large class of materials ranging from classic to exotic semiconductor materials, photovoltaics, thermoelectrics, and metallic systems; we can take advantage of the near-linear scaling of our code up to several thousand nodes.

RESEARCH CHALLENGE

Quantum Monte Carlo (QMC) methods are a suite of tools for direct stochastic solution of the many-body interacting Schrodinger Equation. Although QMC methods are considered to be one of the highest-accuracy, first-principles, materials modeling methods available, and demonstrate a long and distinguished history of benchmark calculations. Their usage for materials design and discovery has historically been limited by their large computational cost. With the high performance computing capabilities of Blue Waters, however, it is now possible to extend this method to the realm of high-throughput materials computation and discovery. The goal of our work, therefore, is to develop the first database of materials computations based on quantum Monte Carlo results. We expect that this database will serve as a shared community resource to accelerate the use of this high-accuracy method and advance the community's knowledge of best practices in the application of QMC to real materials design and discovery.

METHODS & CODES

We have developed an automated workflow that enables high-throughput simulations of solid-state materials using variational and diffusion Monte Carlo. Our open-source framework is available on our GitHub pages [1], and currently utilizes CRYSTAL [2] to generate trial wave functions and QWalk [3] for quantum Monte Carlo simulations.

RESULTS & IMPACT

The prospect of materials design using high performance computation is one of the most exciting for future technologies. However, the properties of modern materials are rather complex, and current numerical methods can fail to describe them quantitatively. Today, QMC is a state-of-the-art suite of tools for high-accuracy ab initio modeling. It shows great promise for high-accuracy materials modeling and is already well established in the physics community, but its application to real materials with chemical identity remains fairly young. Our goal is to accelerate the development of the QMC community's collective knowledge base around the use of this method for predictive modeling of real materials. Ultimately, the impact will be to enable quantum Monte Carlo methods to emerge as a standard component of the computational materials modeling toolkit, enabling unprecedented high-accuracy simulation of complex materials, correlated systems, high-Tc superconductors, and other historically challenging materials.

Accomplishments:

- *Autogen:* The implementation of an automated workflow (Fig. 1a) for carrying out QMC calculations of solid materials on Blue Waters. The workflow was used together with Blue Waters to carry out our automated quantum Monte Carlo calculations of semiconductor materials. It is now available to the public via our GitHub pages.
- *Calculation of Thermodynamic Properties of Wide Class of Challenging Materials:* We were able to use the *Autogen* tools to carry out a large, systematic calculation of the thermodynamic properties and formation enthalpies of a large and varied class of materials (Fig. 1b). Our efforts thus far have focused on the calculation of thermodynamic properties (total energies and formation enthalpies) and the band gaps of a wide class of semiconductor materials spanning from conventional (silicon, etc.) to more exotic materials (wide band gap oxides, correlated systems, photovoltaic thin film materials).
- *The establishment of QMCDB:* Our Mongo database "QMCDB," which includes results and provenance, is described in detail on the cached National Data Service Project wiki page (http://bit.ly/2fgoZB4). This database will serve as a vehicle to quickly overcome the current expertise hurdle and bring the QMC methodology into the standard computational modeling toolkit. While today there are several competing materials databases for density functional theory, there is no existing QMC database. Thus, Blue Waters has provided us the opportunity to establish this tool for the worldwide QMC community. To our knowledge, this comprises the largest QMC data set that has been established to date.

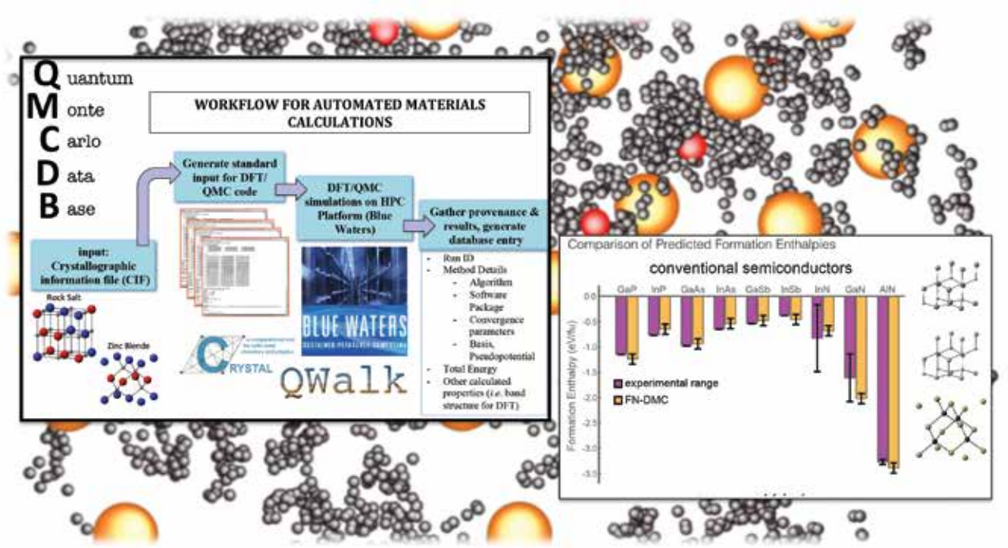

Figure 1: Background: a snapshot of a configuration of electrons around atomic nuclei in the magnesium oxide solid. Foreground: the *Autogen* workflow for automated quantum Monte Carlo calculations of solids and incorporation into quantum Monte Carlo database, and selected results for calculated formation enthalpies of conventional semiconducting solid materials.

WHY BLUE WATERS

Blue Waters is the key to carrying out the comprehensive set of QMC materials calculations that populate our database. QMC methods exhibit near-linear scaling on the entire Blue Waters platform, which has allowed us to calculate properties of an extensive set of materials that would otherwise not be possible. Historically, the wide scale adoption of QMC as a method for materials modeling has been limited by its large computational cost, but Blue Waters allows us a first key opportunity to overcome the computational barrier.

PUBLICATIONS AND DATA SETS

Schiller, J. A., L. K. Wagner, E. Ertekin, Phase Stability and Properties of Manganese Oxide Polymorphs: Assessment and Insights from Diffusion Monte Carlo. *Phys. Rev. B*, 92:23 (2015), DOI: 10.1103/PhysRevB.92.235209.

Yu, J., L. K. Wagner, E. Ertekin, Towards a systematic assessment of errors in diffusion Monte Carlo calculations of semiconductors: case study of zinc selenide and zinc oxide. *J. Chem. Phys.*, 143:22 (2015), DOI: 10.1063/1.4937421.

Yu, J., L. K. Wagner, E. Ertekin, Fixed node diffusion Monte Carlo description of nitrogen defects in zinc oxide. Phys. Rev. B, 95:7 (2017), DOI: 10.1103/*PhysRevB*.95.075209.

Quantum Monte Carlo Database (QMCDB).