# QMCDB: A LIVING DATABASE TO ACCELERATE WORLDWIDE DEVELOPMENT AND USAGE 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 as part of our development of QMCDB (Quantum Monte Carlo DataBase), a database of materials properties calculated via the highly-accurate quantum Monte Carlo technique. The systems simulated are to be incorporated into our database, which will be made available to the international materials modeling community. This platform for easy, searchable data exchange will accelerate the knowledge base around the use of QMC for materials modeling and enable its evolution from a physics approach to a tool for real engineering materials design. 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 methods and code up to several thousand nodes.

## INTRODUCTION

Quantum Monte Carlo (QMC) methods are a suite of tools for direct stochastic solution of the many-body interacting Schrödinger 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 (HPC) 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 & RESULTS

The prospect of materials design using HPC is one of the most exciting for future technologies. This has been already achieved to some degree using established methods on model problems. 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 (model systems, effective Hamiltonians), but its application to real materials with chemical identity remains fairly young. The reason for this is that historically, quantum Monte Carlo methods were perceived as being too computationally costly for wide-scale adoption. Modern implementations of the algorithms, together with the leadership-class computing facilities offered by Blue Waters open a new opportunity in this field.

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. We have used Blue Waters to carry out high-throughput calculations of the properties of a large class of semiconductors and oxides, the results of which are to be incorporated into a newly developed database. 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.

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). 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, **for the first time**.

To date, our work has encompassed:

- The implementation of an automated framework for carrying out QMC calculations of solid materials on Blue Waters. The framework is working and is now being used together with Blue Waters to carry out our automated quantum Monte Carlo calculations of semiconductor materials.
- The establishment of QMCDB: Our MongoDB database "QMCDB" is now active through National Data Service Labs, using cloud capabilities. The database is described in detail on the National Data Service Project Wiki page, at http://wiki.nationaldataservice.org/QMCDBProject. Once we populate the database with our initial set of 20 materials calculations (largest QMC data set to date) with provenance, it will become publicly accessible.

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-$T_c$ superconductors, and other historically challenging 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 this barrier.

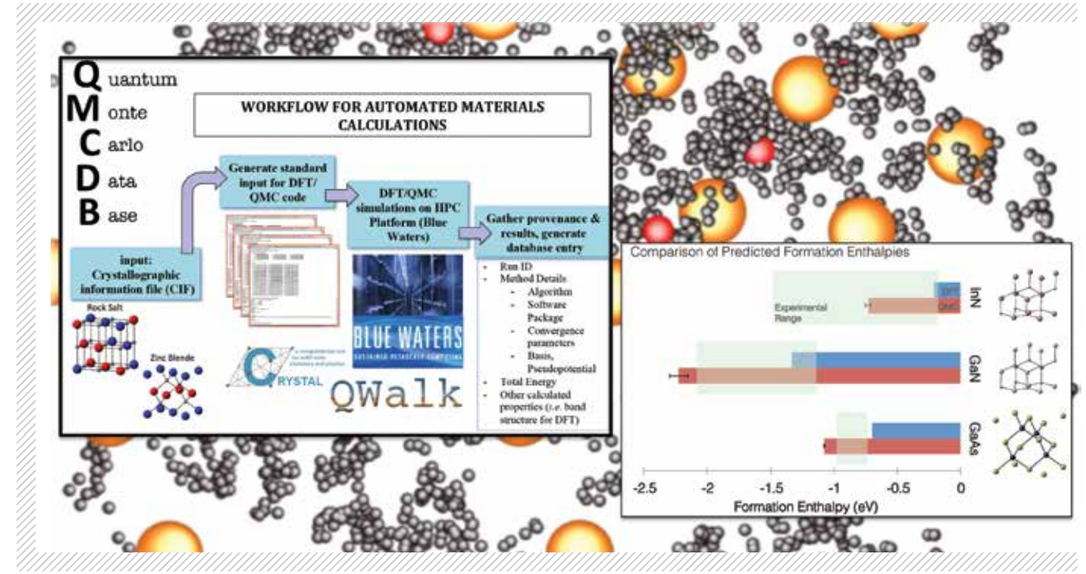


FIGURE 1: A snapshot of a configuration of electrons around ionic nuclei in the magnesium oxide solid. Foreground: workflow for automated quantum Monte Carlo calculations of solids and incorporation into quantum Monte Carlo database, and selected results for calculated formation enthalpies of conventional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

## NEXT GENERATION WORK

Next generation Track-1 systems in the coming years will enable us to carry out more systematic analyses on a larger class of materials. Our initial work shows that simulations of larger sized systems are important. Also, more systematic studies will enable us to carry out big data assessments of correlated many-body wave functions, towards the idea of "correlated electron genomics."

## PUBLICATIONS AND DATA SETS

Schiller, J.A., L.K. Wagner, and E. Ertekin, Phase stability and properties of manganese oxide polymorphs: Assessment and insights from diffusion Monte Carlo, *Phys. Rev. B*, 92 (2015), 235209 doi: 10.1103/PhysRevB.92.235209

Yu, J., L. K. Wagner, and 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 (2015), 224707, doi:10.1063/1.4937421

# COLLABORATIVE RESEARCH: INNOVATIVE *AB INITIO* SYMMETRY-ADAPTED NO-CORE SHELL MODEL FOR ADVANCING FUNDAMENTAL PHYSICS AND ASTROPHYSICS

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## EXECUTIVE SUMMARY

We use Blue Waters to carry out large-scale modeling of light and medium-mass nuclei, including short-lived nuclei not yet accessible to experiment but critical to understanding astrophysical processes, which are the focus of current and next-generation rare isotope experimental facilities. The scale of computational challenges inherent to modeling such intricate quantum many-body systems makes the utilization of Blue Waters resources essential for addressing long-lasting challenges of importance to nuclear theory and experiment, as well as astrophysics. **Breakthrough** theoretical advances [1] coupled with Blue Waters' **cutting-edge** computational power have opened a new region, the intermediate-mass nuclei from fluorine (F) to calcium (Ca) isotopes, for **first** investigations with *ab initio* methods. This breakthrough fundamentally advances our understanding of nucleosynthesis, as nuclear energy spectra and reaction rates for many short-lived nuclei involved in nucleosynthesis are **not yet accessible by experiment or reliably measured** for the astrophysically relevant energy regime.





## INTRODUCTION

One of the quintessential open challenges in contemporary physics is to design a comprehensive many-body theory for modeling and predicting nuclear structure and reactions starting from inter-nucleon forces that are consistent with the underlying theory of Quantum Chromodynamics (QCD). The ultimate goal of *ab initio* theory is to find a solution to this problem, which is a computationally-intensive endeavor due to a dual challenge, namely, the non-perturbative nature of QCD in the low-energy regime and the complexity of many-particle nuclei. Short-lived nuclei, currently **inaccessible to experiment**, are key to understanding processes in extreme environments. From stellar explosions to the interior of nuclear reactors, first-principle nuclear models that hold predictive capabilities have been and will be demonstrating a tremendous impact on advancing our knowledge at the frontiers of astrophysics, neutrino physics, and applied physics.

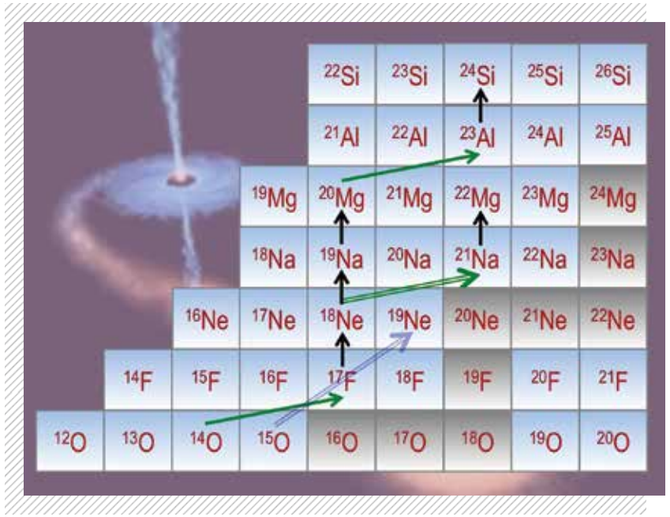


FIGURE 1: Nuclear region of interest to X-ray burst nucleosynthesis—it is now opened up for *ab initio* investigations by the unique capabilities of our SA-NCSM symmetry-guided concept and Blue Waters.

## METHODS & RESULTS

We have developed an innovative *ab initio* nuclear structure approach, symmetry-adapted no-core shell model (SA-NCSM) [1], with concomitant computer code dubbed *LSU3shell* [2], that embraces the first-principles concept and capitalizes on a new symmetry of the nucleus. The *ab initio* SA-NCSM solves the time-independent Schrödinger equation as a Hamiltonian matrix eigenvalue problem. The main computational task is to evaluate a large symmetric Hamiltonian matrix and to obtain the lowest-lying eigenvectors that correspond to the experimental regime. Accuracy is based on the degree of convergence, which is linked to the size of the model space that can be achieved. The SA-NCSM utilizes physically relevant model space of significantly reduced dimensionality compared to ultra-large model spaces encountered by standard *ab initio* approaches. These theoretical advances coupled with the computational power of Blue Waters allow us to reach medium-mass nuclei that are inaccessible experimentally and fall far beyond the limits of other *ab initio* methods. The nuclei of interest represent a considerable challenge requiring computational power of near full-capacity of Blue Waters and its system memory. The following list describes the results and projected studies:

- We have provided the **first** *ab initio* description of the open-shell $^{20}$Ne and $^{18}$F nuclei. Following this success, we target Ne, Mg, and Si isotopes of importance to nuclear reaction studies. Such reactions in the intermediate-mass region are key to further understanding phenomena like X-ray burst nucleosynthesis and the Ne-Na and Mg-Al cycles.
- Work in progress focuses on one of the most challenging problems in nuclear physics today: achieving an *ab initio* description of the so-called Hoyle state in $^{12}$C, the resulting state of the essential stellar triple-alpha process, key to modeling nucleosynthesis and stellar explosions. We have also calculated important low-lying states in $^{12}$C, including negative-parity states and giant monopole and quadrupole resonances.
- We have studied $^{10}$He (the focus of current experimental proposals), with impact on experimental techniques around the neutron-drip line that involve halo nuclei. Further calculations in even larger model spaces, only feasible with Blue Waters, will be key to resolving the observed inconsistency among several experimental results.

## WHY BLUE WATERS

*Ab initio* nuclear structure studies represent an extremely computing-intensive endeavor. To illustrate the level of complexity, applications to medium-mass nuclei require over hundreds of exabytes of memory to store the Hamiltonian