QUANTUM MONTE CARLO SIMULATIONS OF MAGNETISM AND MODELS IN CONDENSED MATTER

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EXECUTIVE SUMMARY

This project uses quantum Monte Carlo techniques to study the behavior of electrons in materials where traditional methods of calculation are too inaccurate to be of use. These large-scale calculations will provide unprecedented detail about the correlated behavior of electrons in these materials at the subnanoscale. The simulations will then be analyzed in a systematic way to extract a compressed description of their behavior, which will help us to understand the difference at a microscopic level between a simple magnetic material and unconventional superconductors.

RESEARCH CHALLENGE

There are special materials in which quantum mechanics are noticeable even at the scale of the entire material. The simplest examples of this are semiconductors, which are by now quite well understood by band structure theory and have had a huge impact on technology. However, materials such as unconventional high-temperature superconductors, magnetic materials, topological materials, and many others have quantum effects that are very challenging to describe under the normal band structure framework. Current tools also struggle to accurately predict the properties even of semiconductors.

The Schrödinger equation, elucidated in the 1920s and carefully tested, tells us how, in principle, to predict the properties of materials. Until recently, however, directly solving the Schrödinger equation for complex materials has been out of reach because of the computational cost.

METHODS & CODES

We use the QWalk package [1], which was developed at the University of Illinois at Urbana-Champaign, to perform quantum Monte Carlo calculations. These methods sample the positions of the electrons using a stochastic process. How often the positions appear in the random sample represents the magnitude of the wave function. In this way, we can incorporate correlation effects between electrons. This method uses a single approximation, called the fixed node approximation, to become efficient. Since there is just one major approximation, it can be improved as more computational resources become available to achieve more and more accurate results.

RESULTS & IMPACT

With Blue Waters (BW), it is now possible to achieve unprecedented accuracy in solving the Schrödinger equation using Monte Carlo techniques. Our group has used BW to show that the simulations represent a step forward in the state of the art in computing many particle quantum systems. We have been able to describe the properties of water interacting with graphene, defects in semiconductors, the properties of superconducting materials, and transitions between metallic and insulating behavior.

The figure shows work performed on BW on vanadium dioxide [2], which transitions from a metal to an insulator at around 340°K (150°F). This transition can also be induced by strain [3] or electric field. For many years, it has been a mystery why this happens, mainly because electron correlation is important in the effect, which is difficult to access with inaccurate quantum calculations. By using BW to perform quantum Monte Carlo calculations of the electrons in this material, we were able to describe the transition from first principles, meaning that we used only the positions of the atoms to compute its properties. This new predictive capability opens the door to computer-based design of these extraordinarily sensitive materials that can be used in new devices.

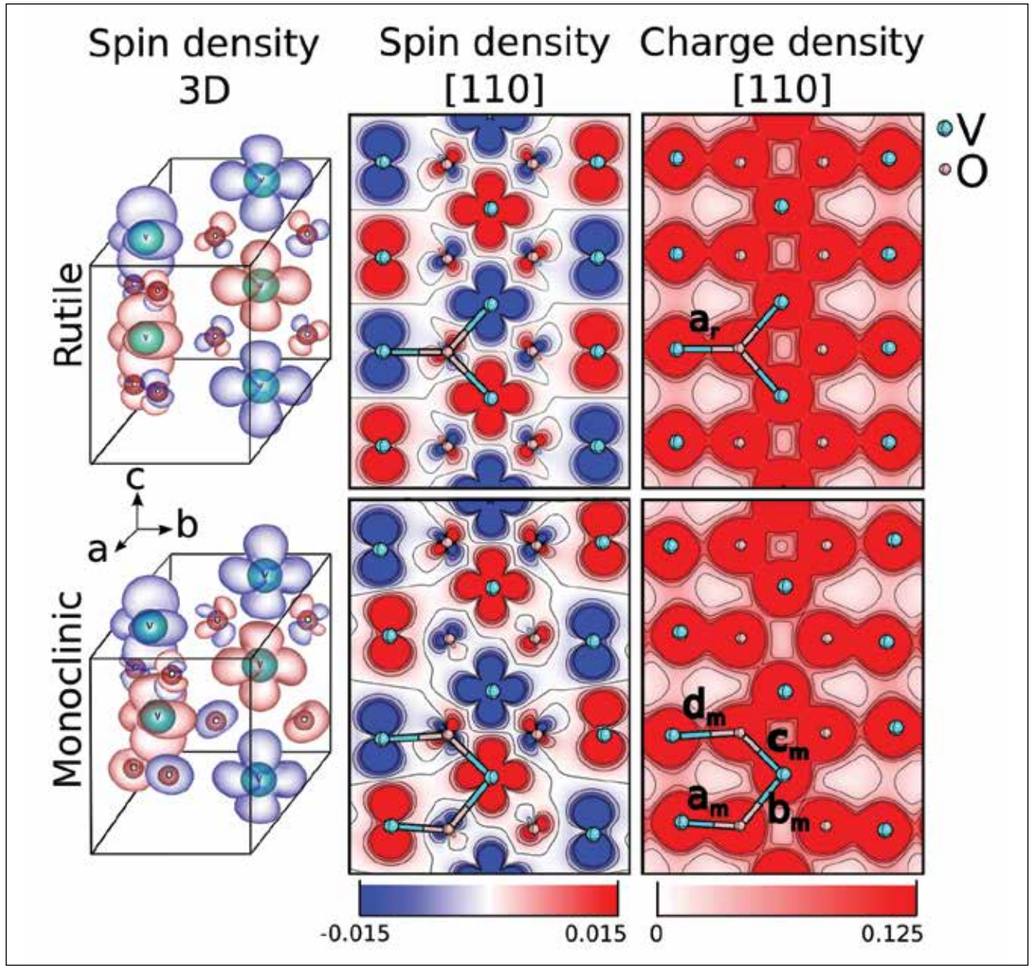
Now that we have accurate calculations available, we would like to learn as much as possible from them. We would like to be able to know how to think about a given material. In physics, this is formally given by using an object known as an effective Hamiltonian, which encodes how a system behaves under experimental conditions. Toward that objective, we are using BW to probe quantum systems and extract effective Hamiltonians by performing rigorous multiscale quantum simulations.

WHY BLUE WATERS

The BW system provides excellent technical support and access to high-performance computing with a minimal amount of effort required from scientists, so that we can focus on the science. As one of the few National Science Foundation-supported resources that provides large allocations, it has been critical to the success of the projects listed here.

PUBLICATIONS AND DATA SETS

Wagner, L.K., and P. Abbamonte, Effect of electron correlation on the electronic structure and spin-lattice coupling of high-*Tc* cuprates: Quantum Monte Carlo calculations. *Physical Review B*, 90 (2014), 125129.



Understanding of a transition of vanadium dioxide from metal to insulator from quantum Monte Carlo (QMC). This material can switch very quickly between these two states, which could enable optical devices and controllably transparent windows. The QMC calculations were the first to successfully describe this transition from the atoms up, without any empirical parameters.

Shiller, J.A., L.K. Wagner, and E. Ertekin, Phase stability and properties of manganese oxide polymorphs: Assessment and insight from diffusion Monte Carlo. *Physical Review B*, 92 (2015), 235209, DOI: 10.1103/PhysRevB.92.235209.

Wu, Y., L.K. Wagner, and N.R. Aluru, The interaction between hexagonal boron nitride and water from first principles. *The Journal of Chemical Physics*, 142 (2015), 234702.

Wu, Y., L.K. Wagner, and N.R. Aluru, Hexagonal boron nitride and water interaction parameters. *The Journal of Chemical Physics*, 144 (2016), 164118.

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. *The Journal of Chemical Physics*, 143 (2015), 224707.

Yu, J., L.K. Wagner, and E. Ertekin, Fixed-node diffusion Monte Carlo description of nitrogen defects in zinc oxide. *Physical Review B*, 95 (2017), 075209.

Zheng, H., and L.K. Wagner, Computation of the correlated metal-insulator transition in vanadium dioxide from first principles. *Physical Review Letters*, 114 (2015), 176401.