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ACCURATE EFFECTIVE INTERACTIONS IN QUANTUM MATERIALS

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EXECUTIVE SUMMARY

In this project, the research team developed multiscale quantum models from first-principles (*ab initio*) calculations that explicitly consider the subatomic physics of electrons and nuclei. The work represents a new level in connection between the subatomic scale and up to nanometer- and micrometer-scale quantum effects. The research is enabled by a combination of high-performance computing to gather data and advanced data science techniques to analyze the resultant data. The ensuing multiscale-effective models may then be used to predict the behavior of materials at those length scales and to understand quantum materials, for which quantum effects are starkly visible in their behavior.

RESEARCH CHALLENGE

All materials that we deal with day to day are made up of the same ingredients—nuclei and electrons. The variety of different objects and devices emerges from the many-particle behavior of these systems. How that actually occurs is still not well understood in many cases. Part of the challenge is that the electrons and nuclei behave according to quantum mechanics, whose equations are notoriously difficult to solve for more than just a few particles. Researchers use the concept of effective models to describe the behavior without having to explicitly consider the details of the fundamental particles. These effective models are used to design devices and otherwise describe how the materials behave. However, for new and emergent materials, it is often not known how to connect these effective models to the underlying dynamics of electrons and how to predict the correct effective model without appealing to experiment.

METHODS & CODES

The research group has recently developed a way of exploring the quantum solution space, which in turn allows them to derive effective models from high-accuracy first-principles calculations. The method involves performing many Monte Carlo calculations to simulate the electrons, which can run in parallel on thousands of nodes, and to analyze their results. The team uses the open-source QWalk [1] code, which was developed at the University of Illinois at Urbana–Champaign.

RESULTS & IMPACT

Fig. 1 shows the application of the effective model ideas to real materials. The question the research group is trying to answer in this study is ultimately why some materials (called unconventional superconductors) superconduct at high temperatures while some do not, which has been an open question in the field for decades.

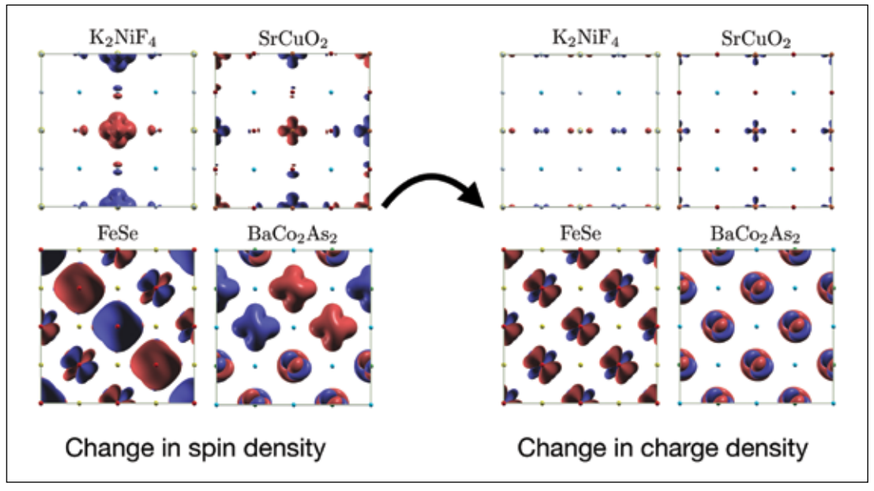

Figure 1: For some materials, spin and charge are linked together; a change in one will lead to a change in the other. This diagram shows that the superconductors $SrCuO_2$, FeSe, and $BaCo_2As_2$ all have coupling between charge and spin, while the simple antiferromagnet K_2NiF_4 does not.

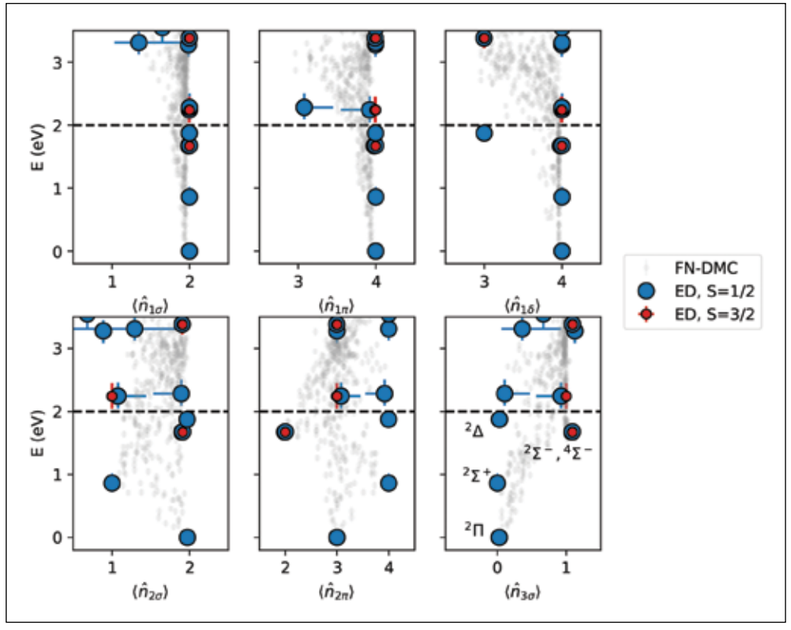

Figure 2: Data analysis of the CuO molecule. The gray dots are the simulation data generated on Blue Waters, while the blue and red dots correspond to the solution of the effective model that is fit to those data. The x-axis quantities are descriptors that are used to describe the effective model. The solution matches the experimental observations of the molecule.

Fig. 2 shows the application of a more general version of the effective model-fitting technique. Again, the team performed highly accurate calculations on a small molecule, CuO, which were then analyzed using statistical techniques to determine an effective model for the system. While it is not possible to solve the full system exactly to find its excitation spectrum, the researchers were able to solve the reduced effective model, which agrees with experiment. In fact, many of the excitations that were found are not easily computed in the full system. During this work, the team was able to dramatically improve the efficiency of their model-generation algorithms because of the large amount of data gathered using Blue Waters.

There have been a number of other projects that have been enabled by this computational resource. These have been performed in close collaboration with experimentalists, and several predictions have inspired new experiments on materials.

WHY BLUE WATERS

Blue Waters was critical for the performance of this work. The quick turnaround of the calculations enabled the research team to accelerate the development of challenging methods by a large, difficult-to-quantify amount. In fact, some of these problems would have been unapproachable without the power of the Blue Waters system.

PUBLICATIONS & DATA SETS

B. Busemeyer, G. J. MacDougall, and L. K. Wagner, "Prediction for the singlet-triplet excitation energy for the spinel $MgTi_2O_4$ using first-principles diffusion Monte Carlo," *Phys. Rev. B*, vol. 99, p. 081118, 2019.

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A. R. Munoz, L. Kish, G. J. MacDougall, and L. K. Wagner, "Benchmarking computed and experimental neutron form factors for MnO and NiO," in preparation, 2019.

J. N. B. Rodrigues and L. K. Wagner, "Charge-spin susceptibility as a useful probe of quantum materials," in submission, 2019.

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