From real materials to model Hamiltonians: multiscale modelling of strongly correlated electronic systems with many body wavefunctions

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Given a realistic material with all its intrinsic complications, how does one develop simple reliable models for understanding its properties? Theoretical insight has been the key driver of this process leading to simple few-band pictures. When the interactions are comparable or much stronger than the kinetic energy, it is convenient to adopt the real space lattice approach. The effective Hamiltonian involves a considerable renormalization of parameters. Estimating these values is important for several applications in physics and chemistry. Bands/states far from the Fermi level play a significant role in the screening of Coulomb interactions leading to Hubbard U's that have been traditionally hard to determine reliably. Here we discuss an approach that we have been developing which builds in several safeguards and provides a way of obtaining effective Hamiltonians which can be simulated using techniques specifically defined for small local Hilbert spaces.

I. INTRODUCTION TO DOWNFOLDING

HJC: Survey of DFT based downfolding and known limitations....Survey of model Hamiltonians - Hubbard, Kanamori, ... HJC: Modify intro - this is from the paper.....

Reliably simulating quantum systems remains a challenge for the physics and chemistry communities. Calculations on real materials are primarily carried out with density functional theory (DFT), which in principle is exact, but in practice is limited by the quality of the exchange functional. On the other hand are approximate model Hamiltonians (such as the Hubbard model) which describe the low energy physics solely in terms of the valence electrons and are crucial to our understanding of physical phenomena such as antiferromagnetism and high temperature super-

conductivity. However, transitioning from actual materials to an effective Hamiltonian on a lattice relies on physical insight and/or fits to experimental data, and this notion is not always rigorously well justified.

This effective reduction of the complexity of the calculation is known as "downfolding" and is commonly carried out using DFT-based approaches. These methods use the electronic band structure to generate a tight binding model, but are generally silent on the role of interactions (i.e. two and higher body terms). Incorrect estimation of the strength of the interactions in the case of systems with strong electron-electron correlations leads to outcomes in clear violations of experiments. For example, the local density approximation (LDA) predicts undoped $LaCu_2O_4$ to be metallic instead of insulating, requiring adhoc inputs for the downfolding procedure. Other schemes for magnetic systems involve fitting DFT energies for various spin configurations to effective Heisenberg and bilinear biquadratic models.

Once an effective Hamiltonian in the *reduced* Hilbert space is obtained, it can be used to perform a lattice-model calculation on a *much* bigger system. Many techniques such as the density matrix renormalization group (DMRG), tensor networks, dynamical field theory (DMFT), density matrix embedding (DMET) and lattice quantum Monte Carlo (QMC) methods have been developed to deal with such model Hamiltonians. This multi-step modeling procedure is needed since the *ab-initio* calculations for a given system size are, in general, much more computationally expensive than the equivalent lattice calculations. Large sizes are crucial to study finite size effects, and in turn theoretically establish the presence of a phase. For example, in the case of frustrated magnetic systems, many nearly-degenerate states exist some of which can be stabilized only on very large unit cells. In addition, excited states and dynamical correlation functions have traditionally been difficult in *ab-initio* approaches, but have seen progress for lattice model methods.

Various efforts have been taken to develop accurate downfolding methods. For example, Aryasetiawan et al. have laid out a post-DFT framework for obtaining static and frequency dependent Hamiltonians using the constrained-RPA approach. While the method has been applied to a wide range of systems, it is only recently that its accuracy is being rigorously checked. Other approaches include traditional Lowdin downfolding within a stochastic approach and canonical transformation theory, also being actively pursued as quantum chemistry methods

In this paper, we apply the *ab-initio* Quantum Monte Carlo (QMC) approach [**?** **?**], one of the most accurate and highly scalable wavefunction based electronic-structure methods. The QMC method works directly in the continuum and explicitly introduces correlation into the Slater de-

terminant obtained from DFT or Hartree Fock calculations. For energies of eigenstates, obtained by stochastic projection, the only systematic error is in the assumption of the nodes of the many-body wavefunction: in practice, this approximation is estimated to miss $5-10\%$ of the correlation energy. Correlation functions and density matrices are generally less accurate, but good wavefunctions can bring down the errors significantly. For arbitrary wavefunctions which can be evaluated in polynomial time, the *only* error is purely statistical and can be brought down to arbitrarily small values by increased sampling.

While all results presented here utilize data generated from *ab-initio* QMC, the idea of fitting data from many-body wavefunctions to models applies to other methods as well. Information from many-body wavefunctions detects inadequacies of a proposed model: the deviations suggest the form of additional missing terms. Crucially, a wavefunction-based method does not differentiate between kinetic and potential energy terms: all terms are treated the same way. The approach is complementary to semi-empirical methods that fit models solely to available energy spectra.

II. CRITERIA FOR HAMILTONIAN MATCHING

HJC: Visualizing the goodness of fit/connecting to tools from data science HJC: Choice of norm and avoiding overfitting: L_1, L_2 or something else?

III. SIMPLE EXAMPLES

HJC: Choosing the optimal one body space/ Dual optimization of Hamiltonian parameters and basis sets HJC: Simple examples - lattice to lattice downfolding - Three to One band model and the "effective" d orbitals

IV. REALISTIC APPLICATIONS - SILICON, CARBON, TRANSITION METALS, TRANSITION METAL OXIDES

V. PROSPECTS

HJC: The need for including spin-orbit terms, need for QMC for this case HJC: Other areas: Magnetism, small energy scales HJC: Applications to non QMC methods - coupled cluster, FCI, HCI HJC: Strengths and limitations of effective Hamiltonian approach