Efficient, Pseudopotential-Free Auxiliary-Field Quantum Monte Carlo Calculations in Solids

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We present an approach for efficient, pseudopotential-free many-body calculations in periodic solids using the phaseless auxiliary-field quantum Monte Carlo (AFQMC) method [1]. We employ the frozen core (FC) technique to obviate the need for pseudopotentials. In parallel to many-body quantum chemistry methods, tightly-bound inner electrons occupy frozen canonical orbitals, which are determined from a lower level of theory, such as Hartree-Fock or CASSCF. Since AFQMC random walks take place in a many-electron Hilbert space spanned by a one-particle basis, FC can be realized without introducing additional approximations. The same formalism also allows a basis transformation (downfolding) to an effective one-particle orbital basis using, for example, a truncated set of Kohn-Sham DFT orbitals. Both FC and downfolding provide significant computational savings over fully correlating all the electrons in full plane-wave basis, while retaining excellent transferability and accuracy. We demonstrate the approach by calculating the equation of state of crystalline MnO in antiferromagnetic and ferromagnetic phases. Twist-average boundary condition [2] and a finite-size correction [3] are employed to minimize the effect of finite simulation cell. AFQMC in this basis reproduces the results of the full plane-wave basis many-body calculations, and leads to accurate determination of the magnetic ordering.

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[1] S. Zhang and H. Krakauer, Phys. Rev. Lett. **90**, 136401 (2003).

[2] C. Lin, F. H. Zong, and D. M. Ceperley, Phys. Rev. E **64**, 016702 (2001).

[3] F. Ma, S. Zhang, and H. Krakauer, Phys. Rev. B **84**, 155130 (2011).

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