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Achieving Chemical Accuracy in Li-rich Layered Oxide Materials via Quantum Monte Carlo Method KAYAHAN SARITAS, ERIC FADEL, Massachusetts Institute of Technology, Department of Materials Science, BORIS KOZINSKY, Bosch Research and Technology Center in North America, JEFFREY C. GROSSMAN, Massachusetts Institute of Technology, Department of Materials Science — Density Functional Theory (DFT) is the most popular and versatile method for atomic scale modeling and design of new cathode materials such as Li-rich layered transition metal (TM) oxides . However, all current DFT functionals fail to accurately model the s-to-d or p-to-d orbital charge transfer energies present in TM atoms. Although DFT can be corrected by empirical parameters, such as Hubbard-U, their transferability on different systems can be questionable. Quantum Monte Carlo (QMC) is the method that treats electrons explicitly to solve the many-body Schrodinger equation exactly. Especially for formation energies of solids, QMC is the only method that can achieve near chemical accuracy while being applicable to systems with 100s of electrons. We discuss our recent application of QMC methods to $LiCoO_2$ and $LiNiO_2$ to understand the bonding mechanisms during charging and discharging. We show that QMC is able to accurately predict the operating voltages and energies of the localized d-states of transition-metal atoms without any empirical parameters. We highlight the possible use of QMC in designing Li-rich layer oxide alloys for future generation cathode materials, and to serve as a critical benchmark for calibrating DFT methods for the accurate materials design.

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