Abstract Submitted
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Modeling of Materials for Energy Storage: A Challenge for Density Functional Theory[1] MERZUK KALTAK, MARIVI FERNANDEZ-SERRA, Department of Physics and Astronomy, Stony Brook University, NY, MARK S. HYBERTSEN, Center for Functional Nanomaterials, Brookhaven National Laboratory, NY — Hollandite α-MnO_2 is a promising material for rechargeable batteries and is studied extensively in the community because of its interesting tunnel structure and the corresponding large capacity for lithium as well as sodium ions. However, the presence of partially reduced Mn ions due to doping with Ag or during lithiation makes hollandite a challenging system for density functional theory and the conventionally employed PBE+U method. A naive attempt to model the ternary system $Li_xAg_yMnO_2$ with density functionals, similar to those employed for the case $y = 0$, fails and predicts a strong monoclinic distortion of the experimentally observed tetragonal unit cell for $Ag_2Mn_8O_{16}$. Structure and binding energies are compared with experimental data and show the importance of van der Waals interactions as well as the necessity for an accurate description of the cooperative Jan-Teller effects for silver hollandite Ag_yMnO_2. Based on these observations a ternary phase diagram is calculated allowing to predict the physical and chemical properties of $Li_xAg_yMnO_2$, such as stable stoichiometries, open circuit voltages, the formation of Ag metal and the structural change during lithiation.

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Merzuk Kaltak
Department of Physics and Astronomy, Stony Brook University, NY

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