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Charge versus orbital-occupancy ordering in manganites WEIDONG LUO, MARIA VARELA, JING TAO, STEPHEN J. PENNYCOOK, SOKRATES T. PANTELIDES, Vanderbilt University and Oak Ridge National Laboratory — It is generally assumed that density-functional theory (DFT) in the local-spin-density approximation (LSDA) or the generalized- gradient approximation (GGA) is not adequate to describe mixed- valence manganites. Here we report benchmark DFT/GGA calculations for the ground-state structural, electronic and magnetic properties for both undoped and doped $\mathrm{CaMnO_3}$ and find the results to be in excellent agreement with available data, including new atomic-resolution Z-contrast imaging and electron-energy loss spectra. More specifically, we found that the DFT results predict two inequivalent Mn atoms in both 0.33 and 0.5 electron-doped $\mathrm{CaMnO_3}$, in agreement with experimental evidence of $\mathrm{Mn^{+3}/Mn^{+4}}$ oxidation state ordering. The inequivalent Mn atoms are marked by their distinctive orbital occupancies, dissimilar local Jahn-Teller distortion and different magnetic moments from DFT calculations. We also show that the spherically integrated charges associated with the two inequivalent Mn atoms are the same, and they are actually the same as in the Mn metal. This charge neutrality with different orbital occupancies is the result of self-consistency and atomic relaxations in the crystal. We conclude that DFT without additional correlations can account for the observed properties of oxidation-state ordering in this system. The impact of the results on other mixed-valence systems will be discussed.

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