Magnetic 3$d$ interactions in ZnO and In$_2$O$_3$ in a band-gap corrected approach

STEPHAN LANY, H. RAEBIGER, A. ZUNGER, Natl. Renewable Energy Lab — The electronic and magnetic configuration of 3$d$ transition metal (TM) impurities in wide-gap oxides like ZnO and In$_2$O$_3$ is misrepresented in the standard LDA and GGA approximations to density functional theory: Because the conduction band minimum lies energetically much too low, the spin-polarized impurity states wrongly occur as resonances inside the conduction band rather than as gap states. Due to spurious “charge spilling” from the TM impurity state into the host conduction band, the magnetic moment and the occupancy of the TM impurity state is incorrect, and the TM state becomes partially occupied, which is prone to cause overestimated ferromagnetic interactions. These errors are not corrected by the LDA+U or GGA+U methods often applied to TM-$d$ states. In our band-gap corrected approach, we augment the GGA+U functional by empirical non-local external potentials (NLEP) for the $s$- and $p$-states of Oxygen and the cations. In this approach the correct spin and orbital configuration of the TM impurity-states is recovered. In the absence of additional doping, we find generally short-ranged magnetic interactions, and pronounced Jahn-Teller effects in case of partially occupied gap states. Additional electron-doping can lead to more long-range ferromagnetic interactions for those TM-dopants that have unoccupied $d$-states which hybridize strongly with the conduction band.

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