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Localization, lattice distortion, charge transition levels, and magnetism of small-polaronic hole- and electron-states in wide-gap semiconductors[1]

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The formation of a small polaron, i.e. of a localized (electron or hole) quasi-particle state that is stabilized by a lattice distortion, is a problem in solid state physics that has eluded a quantitative description by first principles Hamiltonians for a long time. Specifically, conventional density functional theory calculations typically predict a much too delocalized state and usually fail to correctly predict the lattice distortions of localized hole-states in semiconductors and insulators. While this problem has been studied in detail for some prototypical cases like the Al impurity in SiO_2, it has at the same time precluded an extensive theoretical literature on the phenomenology of systems with localized hole states, despite the potentially dramatic effect of hole localization on such timely research topics as p-type doping of oxides or that of diluted magnetic semiconductors. Indeed, many predictions for hole-introducing defects and impurities that were based on local density approximations have led to a qualitatively wrong physical picture about the lattice distortion, the energies of the hole-bearing acceptor levels in the gap, and about ferro-magnetic interactions between defects. In order to stabilize the polaronic localized states in the gap, we define a parameterized hole- (or electron-) state potential which increases the energy splitting between occupied and unoccupied orbitals, where we further require that a fundamental physical condition is satisfied, i.e., the piecewise linearity of the energy as a function of the occupation number. This requirement takes the form of a generalized Koopmans conditions, which uniquely determines the one free parameter of the hole- (electron-) state potential. Applying this method to the anion-p orbitals within the II-VI series of ZnO, ZnS, ZnSe, and ZnTe, we demonstrate electronic correlation effects remove the partial band occupation and the metallic band-structure character that is predicted by local density calculations forcation vacancies in II-VI semiconductors. This transition dramatically changes the structural, electronic and magneticproperties along the entire series and impedes strongly the ferromagnetic coupling between vacancies. Thus, our results demonstrate that important correlation effects due to open$p$ shells exist not only for first-row ($2p$) elements, butalso for much heavier anions like Te ($5p$). We further employ our method to determine the charge transition states caused by acceptors in wide gap semiconductors (ZnO, In_2O_3, SnO_2, GaN), as well as the self-trapped electrons and holes in TiO_2.

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