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Ab Initio **Modeling of Transition-Metal Impurities in MgO**[1]
SERGEY V. LEVCHENKO, SEBASTIAN ALARCON VILLASECA, ALIAKSEI MAZHEIKA, MATTHIAS SCHEFFLER, Fritz-Haber-Institut der MPG, Berlin, DE — Fe- and Ni-doped MgO are promising materials for the catalytic conversion of methane and CO_2. However, theoretical studies of these materials are scarce. The self-interaction error (SIE) in approximate DFT leads to an incorrect description of the electron localization and hybridization between d states of Ni or Fe and the oxide electronic bands. Replacing a fraction α of the (semi-)local exchange by the exact exchange reduces the SIE, but α remains a parameter depending on the target property. We explore the dependence of the formation energies of Ni_{Mg} and Fe_{Mg} substitutional defects in MgO on α in the Heyd-Scuseria-Ernzerhof hybrid functional (HSE), and compare the results to CCSD(T) embedded-cluster calculations. For Ni_{Mg} defects HSE($\alpha = 0.3$) reproduces CCSD(T) formation energies and CO adsorption energies on Ni_{Mg}. However, $\alpha = 0.48$ is needed in the case of Fe_{Mg}. For both Ni_{Mg} and Fe_{Mg}, $\alpha = 0.44$-0.50 satisifies best the exact DFT condition that the HOMO does not depend on occupation. Contrary to PBE and HSE06, HSE($\alpha \approx 0.5$) reproduces the experimentally observed $O_h \rightarrow D_{4h}$ (oblate) Jahn-Teller distortion for Fe_{Mg}.

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