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Effective on-site Coulomb interaction and electron configurations in transition-metal complexes from constraint density functional theory KENJI NAWA, KOHJI NAKAMURA, TORU AKIYAMA, TOMONORI ITO, Mie University, MICHAEL WEINERT, University of Wisconsin-Milwaukee — Effective on-site Coulomb interactions (U_{eff}) and electron configurations in the localized d and f orbitals of metal complexes in transition-metal oxides and organometallic molecules, play a key role in the first-principles search for the true ground-state. However, wide ranges of values in the U_{eff} parameter of a material, even in the same ionic state, are often reported. Here, we revisit this issue from constraint density functional theory (DFT) by using the full-potential linearized augmented plane wave method. The U_{eff} parameters for prototypical transition-metal oxides, TMO (TM=Mn, Fe, Co, Ni), were calculated by the second derivative of the total energy functional with respect to the d occupation numbers inside the muffin-tin (MT) spheres as a function of the sphere radius. We find that the calculated U_{eff} values depend significantly on the MT radius, with a variation of more than 3 eV when the MT radius changes from 2.0 to 2.7 a.u., but importantly an identical valence band structure can be produced in all the cases, with an approximate scaling of U_{eff}. This indicates that a simple transferability of the U_{eff} value among different calculation methods is not allowed. We further extend the constraint DFT to treat various electron configurations of the localized d-orbitals in organometallic molecules, $TMCp_2$ (TM=Cr, Mn, Fe, Co, Ni), and find that the calculated U_{eff} values can reproduce the experimentally determined ground-state electron configurations.

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