Toward a first-principles theory of rare-earth ions in crystals

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Density functional theory (DFT), including its extensions designed to treat strongly correlated localized electron systems such as DFT+U and DFT+dynamical mean field theory, has proven exceedingly useful in studying the magnetic properties of solids. However, materials with rare earths (R) have remained a notable exception. The most vital rare-earth magnetic properties, such as magnetocrystalline anisotropy (MA), have been notoriously elusive due to the ubiquitous self-interaction error present in nearly all available DFT flavors. In this work [1], we show explicitly how the orbital dependence of self-interaction error may contradict Hund's rules and plague MA calculations, and how analyzing DFT metastable states that respect Hund's rules can alleviate the problem. We systematically investigate and discuss several rare-earth-containing families, RCo_5, $R_2Fe_{14}B$, RFe_{12}, and RMn_6Sn_6, to benchmark the MA calculations in DFT+U. For all compounds we investigated, we found that our methodology reproduces the magnetic easy-axis, easy-plane, and non-trivial easy-cone anisotropies in full agreement with low-temperature experimental measurements. Besides the fully-numerical ab initio approach, we further illustrate an efficient semi-analytical perturbation method that treats the crystal field as a perturbation in the limit of large spin-orbit coupling. This approach evaluates the rare-earth anisotropy by assessing the dependence of crystal-field energy on spin-quantization axis rotation using 4f crystal-field levels obtained from non-spin-orbit calculations. Our analytical method provides a quantitative microscopic understanding of the factors that control MA and can be used for predicting new high-MA materials. Finally, in addition to bulk materials, we explore the potential of utilizing rare-earth MA in 2D materials.

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