Computationally Driven Evaluation of Magnetocrystalline Anisotropy

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Custom-tailored magnetic materials are a crucial component in numerous modern technologies from data storage to energy conversion and communication technologies. The experimental search for new high-performance magnets can profit considerably from guidance by theoretical simulations. Computational screening approaches however depend on reliable first-principle methods for calculating the key physical properties which underlie the magnetism of the material. One of them is the magnetocrystalline anisotropy (MCA), which contributes to the magnetic hardness. From an atomistic point of view, the MCA defines the alignment of the atomic magnetic moments with respect to the lattice based on the spin-orbit coupling (SOC) between the potential energy landscape and the electron spin. The determination of MCA is a challenging task for both experimentalists and theoreticians, as it is highly sensitive to a number of factors and parameters.

For the well-studied $X(acac)_3$ (X = V, Cr, Mn, Fe) coordination complexes we specifically explore and compare different methods for calculating the MCA: the finite energy difference approach (force theorem) within periodic boundary conditions by means of VASP and all-electron, full-potential perturbative approaches in ORCA ranging from PT2 on PBE DFT to CASSCF-PT2 and NEVPT2. We explicitly study the influence of cluster geometry and various computational parameters at different levels of theory. Finally, we address the challenges involved in high-throughput MCA calculations for bulk materials by discussing the example of $L1_0$-FePt.

References:

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