Accurate DFT estimates of magnetic couplings in chromium-based molecular rings

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Estimating accurately the magnetic couplings from first principle calculations has proven to be difficult despite a lot of effort [1]. Here we present significant progress in this area for three extensively studied prototypical molecular Cr7M (M=Cr, Ni, Zn) rings, using an optimally-tuned range-separated hybrid functional that was shown to provide an accurate description of the electronic structure in a variety of more simple molecular systems [2]. We show that improving the overall description of frontier orbitals, especially with respect to a balanced description of localized and delocalized states, as well as taking into account the structure optimizationwe, we get magnetic coupling values distinguished by a unique site distribution along the heterometallic ring. Using the couplings calculated within DFT as the input parameters for the spin model simulations, we obtain excellent agreement with magnetic susceptibitlity measurements performed for all molecules considered.

References:

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