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A DFT+DMFT study of spin crossover molecules JIA CHEN, CHRIS MARIANETTI, ANDREW MILLIS, Columbia University — Recent studies have shown promise in applying dynamical mean-field theory (DMFT) based methods to molecular systems. Molecules with transition metals are an ideal target system as they may be complex enough to preclude a treatment via accurate quantum chemical methods, yet exhibit sophisticated many-body behaviour which eludes density functional theory (DFT). Here we apply DFT+DMFT to the molecular complex Fe(phen)2(NCS)2 which exhibits a spin crossover transition as a function of temperature. First, we show that DFT overestimates the stability of low spin state and hence yields an excessively large crossover temperature, in agreement with existing DFT studies. Subsequently, we present both DFT+U and DFT+DFMT to show the effect of treating local correlations. When using the standard double-counting and an on-site Coulomb repulsion computed within linear response, we show that both methods excessively favour the high-spin state. Alternate approaches to the methodology are discussed to remedy this deficiency.

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