DFT-based modeling of Molecular Magnetic Materials and Transport

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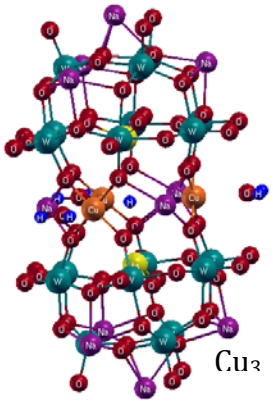
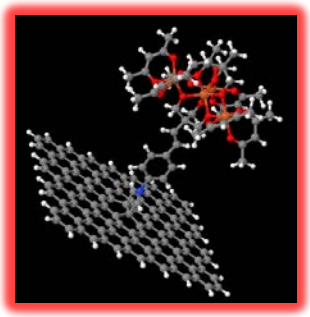


Due to experiments by Sessoli *et al* and Friedman *et al* (1996) there has been a significant amount of experimental and theoretical interest in understanding the behavior of a class of molecules known as molecular magnets. These ~100-atom molecules are typically composed of ~10 transition metal cations that are held in place by ligands. More recently Ishikawa *et al* (2004) have constructed molecular magnets with single lanthanide ions sandwiched between two phthalocyanine molecules. The PBE-GGA has been rather successful in accurately predicting many-of the molecular magnets composed of 3d-transition metal ions. Predictive successes include quantitative determination of magnetic reorientation barriers and the qualitatively correct description of the spin-excitation spectrum. In addition to highlighting these successes, this talk will primarily focuson identifying challenges within this field where self-interaction corrected versions of density-functional theory could lead to better predictive capabilities. For example for the "Cu_3" (above) and "V_{15}" molecular magnets, both of which simplify to a frustrated equilateral triangle of three spin ½ transition-metal cations, the PBE-GGA provides the correct low-energy spin states but overestimates the splittings between the low-lying Kramer doublets and the upper quartet due to the slightly delocalized d-electrons on the transition-metal sites[1]. Improvements in the doublet-quartet splitting due to a more accurate treatment of the coulomb interaction will be discussed [2]. Another challenge, related to understanding spin-dependent electron transfer across molecular magnets, requires one to accurately calculate the charge states of a molecular magnet that is tethered to a distant electrode by a polymer (right). Through applications of DFT involving the Mn_{12} and Fe_4 molecular magnets as possible circuit elements and gold- and spin-polarized graphene flakes (right) as possible substrates, the relation of the level alignment problem to the self-interaction correction will be highlighted. If time permits, a new method for efficiently and exactly accounting for the self-interaction corrections for all electrons will be briefly introduced and discussed[3].



[1]*First-principles study of the spin-electric coupling in a Cu_3 single molecular magnet*, M. F. Islam, JF Nossa, CM Canali and MR Pederson, Phys. Rev. B **82**, 155446 (2010)]; *First-principles studies of the spin-orbit and Dzyaloshinskii-Moriya interactions in the Cu_3 single-molecule magnet*, JF Nossa, MF Islam, CM Canali and MR Pederson, PRB **85** 085427 (2012); *Electric control of a {Fe_4} single-molecule magnet in a single electron transistor*, JF Nossa, MF Islam, CM Canali and MR Pederson, Phys. Rev. B **88**, 224423 (2013).

[2]G. Li Manni and M. R. Pederson.

[3] M. R. Pederson, A. Ruzsinszky, and J. P. Perdew, *A Fast Self-Interaction Corrected (FSIC) Density-Functional with Applications to Atoms and Molecules* (Submitted).