

Theoretical Challenges in Molecular Magnetism and Spintronics

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The goal of this presentation is to give an overview of the accuracy of theoretical methods to calculate magnetic and transport properties in magnetic systems. At single- molecule level, the main key magnitudes are: (i) the exchange interactions between paramagnetic centers; (ii) zero-field splitting parameters (or g tensor) to quantify the magnetic anisotropy; (iii) energy differences between the low- and high-spin states for spin-crossover molecules and transition temperatures. DFT methods provide an accurate description of the exchange interactions for most of the systems. CASSCF- type methods are a very useful tool to determine the local magnetic anisotropy for transition metal and lanthanide systems. For spin-crossover systems, DFT methods can provide a semiquantitative estimation of the low- and high-spin energy differences but post Hartree-Fock methods usually overestimate the stability of the high-spin state. The prediction of transition temperature is beyond the limit of the accuracy of the available methods. Spintronic devices are usually studied by using DFT-NEGF (Non-Equilibrium Green Function) methods that provide a reasonable accuracy of the conductance values and the description of the transport mechanism but also fail in the description of magnetoresistance effects. DFT-NEGF assumes a non-interacting nature of the electrons and employs an equilibrium description using the ground state of the molecule only to analyze a non-equilibrium property, namely, charge transport in a molecular junction.