Magnetic Interactions in Transition Metal Complexes from First-Principles

In this talk I will review our current efforts for the characterization of magnetic interactions in transition metal complexes from first-principles. Since for systems of practical interest, typically containing hundreds of atoms, density functional theory (DFT) is the only alternative for such calculations, I will first focus on the performance of different DFT approximations including a variety of modern density functionals. I will show that hybrid density functionals containing approximately 30% Hartree-Fock type exchange are in general among the best choice in terms of accuracy when compared to available experimental data.[1,2] I will also present a new computational method to evaluate exchange coupling parameters using analytic linear response theory. With this method we avoid the explicit evaluation of energy differences between different broken-symmetry spin multiplets, which typically becomes impractical for large systems. This new method is based on the evaluation of the magnetic torque between two magnetic centers for a given spin configuration using explicit constraints of the local magnetization direction via Lagrange multipliers[3] and it is applicable in combination with any modern density functional with a noncollinear spin generalization. I will show proof-of-concept calculations in small test systems, and in spin frustrated Fe$^{III}$, disk-shaped clusters.[4] Last, I will show our recent efforts for modeling spin dynamics using explicitly real-time evolution in the time-dependent DFT formalism.