Magnetic Exchange Couplings in Transition Metal Complexes from First-Principles Calculations

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In this talk I will review our current efforts for the calculation of magnetic exchange couplings in transition metal complexes from first-principles. For systems of practical interest, typically containing hundreds of atoms, density functional theory (DFT) is the only alternative for such calculations. I will focus on the performance of different DFT approximations including a variety of hybrid density functionals, and show that hybrid density functionals containing approximately 30% Hartree-Fock type exchange are in general among the best choice in terms of accuracy.[1,2] I will also describe a novel computational method to evaluate exchange coupling parameters using analytic self-consistent linear response theory. This method avoids the explicit evaluation of energy differences, which can become impractical for large systems. Our approach 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] This method is applicable in combination with any modern density functional with a noncollinear spin generalization and can be utilized in a “black-box” fashion. I will show proof-of-concept calculations in small test systems, and in frustrated Fe^{III}_7 disk-shaped clusters and dinuclear Cu^{II} complexes.[4,5]

[1] J.J. Phillips and J.E. Peralta, J. Chem. Phys. **2011**, 134, 034108
[2] J.J. Phillips and J.E. Peralta, J. Chem. Theory Comput. **2012**, 8, 3147
[3] J.J. Phillips and J.E. Peralta, J. Chem. Phys. **2013**, 138, 174115
[4] J.J. Phillips, J.E. Peralta, and G. Christou, J. Chem. Theory Comput. **2013**, 9, 5585
[5] J.J. Phillips and J.E. Peralta, J. Phys. Chem. A **2014** (Article ASAP)