Abstract

Our research is focused on the application of *ab initio* (DFT) methods for calculating magnetic properties of open-shell transition-metal complexes, in particular magnetic exchange coupling parameters. The magnetic exchange coupling parameter, J_{ij}, describes the important isotropic spin-spin interactions which dominate magnetic ordering in molecules and solids. The accurate calculation of exchange coupling parameters remains a challenge for DFT methods for primarily two reasons: first, DFT calculations with single Slater determinants struggle to describe multireference spin-eigenstates, forcing one to resort to the use of "broken symmetry" solutions, and secondly, density functionals belonging to rungs 1-3 in "Jacob's Ladder" typically dramatically overpredict the experimental values. Here we report on recent investigations, such as the importance of range-separation of the exchange for correcting the problem of overprediction, the application of the recent Rung 3.5 functionals of B.G. Janesko for describing magnetism in binuclear transition-metal complexes, and the application of Coupled-Perturbed and Constrained DFT methods for calculating coupling parameters.