# Electronic Structure of Molecular Magnets

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Potential implications for quantum computing and low-energy classical storage have recently spurred interest in the development of new quantum memory devices. This has placed new importance on the understanding of fundamental mechanisms underlying molecular magnetism. Magnetism in general is typically conceptualized in the framework of the one-dimensional Heisenberg model. However, the quantitative description for molecular systems requires a balanced treatment involving both static and dynamic correlation effects. Furthermore, experimental exchange coupling constants of molecular magnets are frequently found to be on the order of only a few wavenumbers. This is equivalent to an energy difference between triplet and singlet state of just a few tens of $\mu$Hartree and therefore requires highly accurate predictions for comparison. A standard approach to estimate exchange couplings from DFT alone was proposed by Noodleman [1] and computes the J value from the restricted open-shell Kohn–Sham (ROKS) energy of the high-spin state and the broken-symmetry unrestricted Kohn–Sham (BUKS) energy of the low-spin state. We contrast this with an approach in which the static correlation is treated by the complete active space self-consistent field (CASSCF) method while dynamic correlation is added in the form of n-electron valence state perturbation theory (NEVPT2). For more complicated topologies, especially larger systems exhibiting frustration, already the deterministic evaluation of the Heisenberg model poses challenges. This is especially relevant, as experimental exchange coupling constants are typically obtained by fitting experimental data to these solutions. We describe a path towards solving the Heisenberg model of giant keplerates like $Mo_{72}Fe_{30}$ [2] via projected entangled pair states.

[1]  Noodleman, L. and Davidson, E. R. *Chem. Phys.* **109**, 131 (1986).

[2]  Müller, A., Sarkar, S., Shah, S., *et al.*, *Angew. Chem. Int. Ed.* **38**, 3238 (1999).