Magnetic Properties from Fully Relativistic Theory: Zero-field Splitting and the Pseudospin Hamiltonian

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In this work, we present a method for computation of zero-field splitting parameters of open-shell molecules containing heavy elements. We make use of fully relativistic wavefunction-based methods capable of capturing strong spin-orbit coupling simultaneously with static and dynamical electron correlation. Direct spin-spin coupling is naturally included through the Gaunt interaction, while density fitting ensures computational efficiency. We discuss the construction of a model pseudospin Hamiltonian from the *ab initio* molecular wavefunctions and present preliminary results for zero-field splitting parameter computations. Combining this with our previous work on the response to external magnetic fields and a future extension to account for nuclear magnetic moments, we seek to guide the rational design of new molecular magnets by using these methods for simulation of electron paramagnetic resonance spectra as well as assessment of magnetic anisotropy.