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From Molecules to Materials: Effective Hamiltonians for Spin Dynamics and Magnetic Exchange

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Complex molecular and materials systems often involve multiple interacting electronic and spin degrees of freedom, making the connection between electronic structure and experimental observables challenging. While electronic-structure calculations provide access to the underlying states and interactions, their dynamics and collective behavior often become tractable only when this information is mapped onto suitable effective Hamiltonians. In this talk, I will illustrate how such Hamiltonians can be exploited to describe distinct manifestations of electronic and spin coupling, in molecules and materials. Specifically, I will discuss the photoinduced spin crossover of the open-shell Fe(III) complex $[Fe(qsal)_2]NCS$ and the spin interactions in triangular $Fe(III)_3O$-based metal–organic frameworks. In the first case, a linear vibronic coupling Hamiltonian parametrized from multistate RASSCF calculations enables nonadiabatic dynamics including spin–orbit coupling, providing insight into the coupled electronic and nuclear dynamics underlying ultrafast spin crossover. In the second, broken-symmetry DFT combined with spin-Hamiltonian reconstruction is used to unravel the origin and consequences of magnetic frustration. Finally, I will discuss ongoing efforts to extend nonadiabatic molecular dynamics from molecules to periodic materials.