Spin-vibrational relaxation dynamics in molecular systems

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The interaction of electronic spin and vibrational degrees of freedom mediated by spin-orbit coupling governs spin relaxation in molecular qubits. I derive an effective spin-vibrational Hamiltonian and investigate the temperature and magnetic-field dependence of the leading order spin-vibrational interaction contributions. This analysis reveals previously unexplored, effective interactions with vibrationally non-adiabatic origin that determine the spin relaxation dynamics in the absence of magnetic field. I apply the new approach to simulate spin relaxation of molecular qubits and study the influence of temperature, magnetic field, and symmetry on the coherence properties of these systems. The combination of method development and application offers new insights into the quantum dynamics of spin qubits and enables the engineering of molecular systems for room temperature quantum technology applications.