

Connecting Molecular Electronic Structure and Electron Spin Relaxation for Quantum Information Science

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Quantum devices based on molecules afford unique potential in miniaturization, spatial localization, and tunability through the methods of synthetic chemistry. Unpaired electrons on a molecule constitute a platform for implementing quantum bits (qubits), the fundamental unit of quantum information, as well as molecular quantum sensors (qusors). Spin relaxation destroys the quantum coherences needed to process information, while also providing a potential mechanism for quantum sensing. Thus, understanding spin relaxation constitutes a critical barrier to attaining room temperature quantum applications. While relaxation times have been rationalized on the basis of the Debye model, the assumptions behind this model are incompatible with the structure of molecular materials and fail to yield meaningful predictions for slowly-relaxing, highly coherent molecules. This talk will describe the development of ligand field electron spin dynamics, which has provided a molecular paradigm to quantitatively evaluate and understand temperature- and orientation-dependent electron spin relaxation. These combined experimental and theoretical analyses provide spin relaxation structure-function relationships and elucidate the critical chemical bonding, symmetry, and ligand field and vibronic excited state coupling factors leading to room temperature coherence.