Title: **Single Molecule Magnets for Quantum Computation: Insights from Micro-SQUID-EPR**

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Abstract:

Single-molecule magnets (SMMs) possess robust quantum states that can be reproducibly prepared and tuned by engineering magnetic anisotropy at the molecular level. These systems can display ultraslow relaxation [1] and resonant quantum tunneling of magnetization (QTM), enabling diverse quantum applications [2]- from quantum sensing and quantum memory to multilevel qudits and fault-tolerant quantum computation. Hyperfine levels detected via micro-SQUID (M(H)) measurements and single-electron-spin-transistors have facilitated nuclear spin-based quantum computation [3], while long decoherence times observed in single-ion magnets [4] have renewed interest in these platforms. A complete mapping of the spin manifold - achieved via orientation- and frequency-dependent scans - is essential for integrating SMMs into quantum technologies. The combined micro-SQUID-EPR technique [5] uniquely probes spin states by exciting single crystals with microwaves while simultaneously measuring magnetization. Using this method, M(H) loops recorded across 30 mK - 5 K under variable microwave frequencies reveal absorption peaks corresponding to specific spin transitions. Plotting these peak positions against RF frequencies (0.1 - 40 GHz) and field orientations enables full reconstruction of the spin Hamiltonian [5] and allows direct observation of anti-level crossings. This capability has recently provided a spectroscopic route to visualize topologically quenched tunneling - an expression of geometric phase effects in spin systems—advancing prospects for holonomic, fault-tolerant quantum computation using 4f-based SMMs.

References: 1. J. Am. Chem. Soc. (10.1021/jacs.5c12742) (2025).

2. Chem. Soc. Rev. 47, 501 (2018).

3. Phys. Rev. Lett. 119, 187702 (2017).

4. Nature Chemistry 11, 301 (2019).

5. Nat. Commun. 14, 3361 (2023); doi: 10.1039/D5QI01387A.