Spin-Optical Dynamics in Cr^{3+} Molecular Qubits

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Abstract: We report here the first W-band EPR (95 GHz) study of the spin coherent properties of $Cr(acac)_3$, coupled with its ODMR characterization.

Mononuclear chromium(III) complexes have been shown to be promising Molecular Qubits, where the advantage is the presence of four levels that can be used to generate coherent superposition state. In addition to this, chromium(III) molecular analogues of ruby are particularly attractive, since one can retain the ion's electronic structure while imbuing the system with tunability, portability, and scalability proposed to serve as optically addressable Molecular Qubits.[1] However, there remains a lack of information on the combined optical and spin relaxation properties of Cr(III) molecular complexes, with some notable exceptions.[2]

To begin filling this gap, we report here a combined W-band EPR (94 GHz) and optical study on $Cr(acac)_3$: the use of the high frequency/high field instrument allowed to obtain precise determination of the zero-field splitting parameters and the of the low-lying energy levels in this system. Echo-detected field swept spectrum, reported here for the first time at high field/frequency, indicates that the molecule possesses a short but measurable coherence at low temperature. Both spin lattice relaxation and spin coherence time (Tm) have been measured at different field resonant position at 5 K, evidencing a non-trivial dependence on the transition fields. The possibility of preparing the system in any possible superposition state has been confirmed by performing nutation experiments at different fields. In particular, the remarkably different frequency obtained for low fields (1000 mT) and high fields (3000 mT) indicate that at the former fields the involved transition is an essentially forbidden (3/2→-3/2), while it is a standard (1/2→-1/2) in the latter; this highlight the potential of Cr(III) based systems as multi-level qubits.

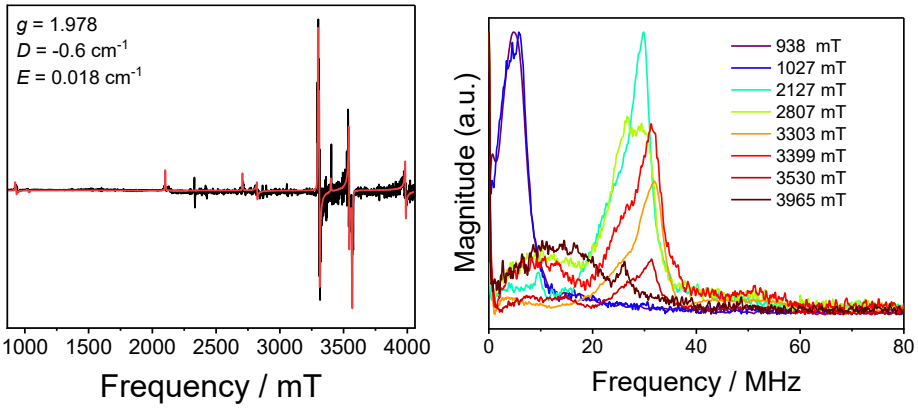

Fig. 1 Left: Continuous wave W-band (94 GHz) EPR spectrum of a powder sample of $Cr(acac)_3$ diluted in the isostructural diamagnetic $Ga(acac)_3$ (1%). Black: experimental spectrum; Red: Simulation with reported parameters. Right: Fourier transform of the nutation trace obtained at different fields.

References

[1] Wojnar, M. K.; Johnson, D. A.; Badding, C. K.; Quan, Y.; Dzikovski, B.; Freedman, D. E. "Crystal Symmetry-Driven Spin-Optical Dynamics in Cr3+ Molecular Spins," Inorg. Chem. **65**, 2884–2893 (2026)

[2] Lenz, S.; Bamberger, H.; Hallmen, P. P.; Thiebes, Y.; Otto, S.; Heinze, K.; van Slageren, J. "Chromium(III)-based potential molecular quantum bits with long coherence times" Phys. Chem. Chem. Phys., **21**, 6976—6983 (2019)