Pulsed and cw High-field EPR Studies on Mn_3^{III} Single-Molecule Magnets

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Introduction

The goal of these experiments was two-fold: (1) to look for spin echoes in a single crystal of a highly anisotropic and well-behaved $S = 6$ $[NEt_4]_3[Mn_3Zn_2(salox)_3O(N_3)_6Cl_2]$ SMM and (2) to investigate the HFEPR spectra for chemically related complexes that have lower symmetry or spin frustrated ground states.

Experimental

Experiments were performed between 3 K and 20 K on single-crystals and ground microcrystalline samples using the Heterodyne and Homodyne EPR spectrometers at NHMFL, respectively. Quasi-optical single-crystal experiments were performed up to 7 T at 240 GHz and 336 GHz, while microcrystalline measurements were taken at 200-435 GHz at fields up to 15 T. Single crystals were aligned such that the molecular easy axis was coincident with the direction of the applied field. Powder samples were finely ground and embedded in eicosane to prevent field-induced torquing of the microcrystallites.

Results and Discussion

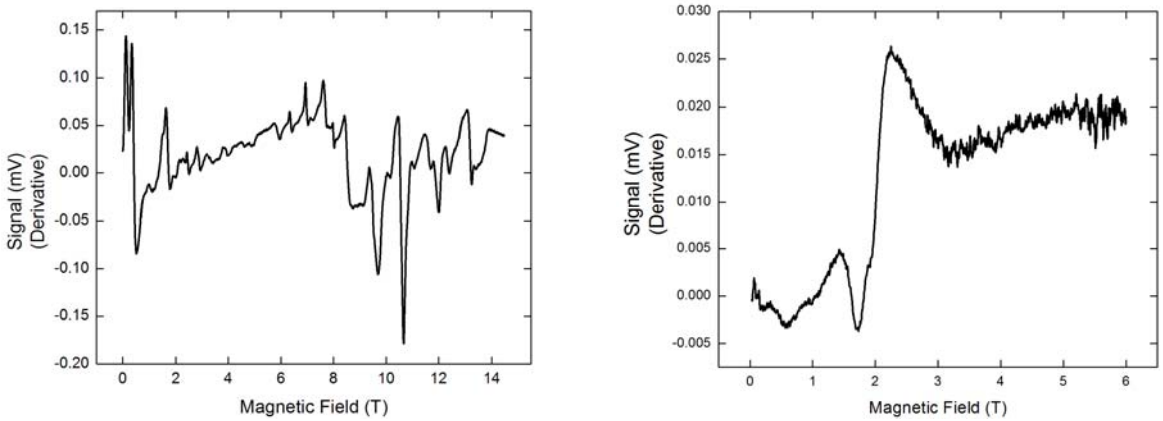

Figure 1: (Left) Powder HFEPR spectrum for $[NEt_4]_3[Mn_3Zn_2(Et\text{-}salox)_3O(N_3)_6Br_2]$, at 10 K and 224 GHz. The plot reveals the large anisotropy and complicated absorption spectrum associated with the two unique molecules in the unit cell. (Right) Powder spectrum for a spin-frustrated analog; the broad/weak absorption is related to the highly spin-mixed ground state.

Conclusions

Although no spin-echoes were observed in the pulsed studies, this result suggests the possible significance of nuclear hyperfine interactions upon the rate of quantum decoherence. To date, no manganese-containing SMMs ($I = 5/2$) have been shown to produce spin-echoes (on the nanosecond timescale), while iron-based SMMs ($I \approx 0$) have yielded spin-decoherence times (T_2) up to 700 ns.[1] Powder HFEPR studies on low-symmetry $S = 6$ complexes reveal a similar axial anisotropy when compared to C_3- symmetric analogs. Since the ac susceptibility and magnetization hysteresis relaxation behavior differ between these complexes, this result provides insight into the quantum tunneling of the magnetization in these complexes.

Acknowledgements

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References

[1] Takahashi, S., *et al.*, arXiv:0810.1254v1 (2008).