Magnetic Response of MnF(salen) at Low Temperatures and in High Magnetic Fields

J.-H. Park, C.C. Beedle (NHMFL); O.N. Risset, D.R. Talham (UF, Chemistry); M. Shiddiq, S. Hill (FSU, Physics and NHMFL); M.K. Peprah, E.S. Knowles, Y.M. Calm, M.F. Dumont, M.W. Meisel (UF, Physics and NHMFL)

Introduction

Linear-chain, integer-spin antiferromagnets, commonly known as Haldane systems, have a storied history in the field of quantum spins, with S = 1 materials receiving the most attention [1]. The S = 2 systems have received less consideration for several reasons, including a smaller Haldane gap, since $\Delta_{S=1} = 0.41\ J$ and $\Delta_{S=2} = 0.09\ J$, where J is the nearest-neighbor exchange energy [2]. Due to a report suggesting Mn(III)F(salen) is a S = 2 Haldane system with $J/k_B \approx 50$ K and no evidence of long-range order down to 2 K [3], we have studied the magnetic response of single-crystals down to 20 mK and in magnetic field up to 18 T.

Experimental Details

Synthesis, XRD, and magnetic (down to 2 K) characterization were performed at UF. The NHMFL EPR facilities were used to study single crystals near 200 GHz, at 4 K, and in fields up to 5 T. The SCM1 station of the NHMFL Millikelvin facility was used to perform magnetometry studies on single crystals down to 20 mK and in fields up to 18 T.

Results and Discussion

The low-field, high-temperature magnetic studies confirmed the results of the initial report [3], and with the use of a home-made pressure cell, the magnetic response was unchanged for P < 1.0 GPa. In addition, several attempts were made to observe ESR lines in a couple of different crystals, and no resonance lines were unambiguously resolved.

On the other hand, a reproducible feature in the response of a torque magnetometer was detected, and the temperature dependence of this feature suggests a critical field of 3.8 T, see **Figure 1**. It is tempting to associate this critical field with the possible closing of a Haldane gap, since the value is consistent with theoretical expectation for $J \approx 50$ K. However, neutron scattering data acquired to date [4] are not consistent with a uniform system containing S = 2 Heisenberg antiferromagnetic chains.

Summary

The nature of the critical field is not yet clear. There are three distinct Mn sites per unit cell, and the motif along the chain is … – 1 – 2 – 3 – 3 – 2 – 1 – 1 – 2 – 3 – 3 – 2 – 1 – … , with three different bridging Mn – F – Mn angles, i.e. 150.4^o (Mn2 – F3 – Mn3), 151.7^o (Mn1 – F2 – Mn2), and 180.0^o (Mn1 – F1 – Mn1 and Mn3 – F4 – Mn3) [3]. In other words, a “trimer-like” arrangement along the chain is suggested, and work is ongoing.

Acknowledgements

This work was supported, in part, by the NSF through DMR-0804408 (SH), DMR-1005581 (DRT), DMR-1202033 (MWM), and DMR-0654118 (NHMFL).

References

[1] Yamashita, M., *et al*., Coord. Chem. Rev. **198**, 347 (2000).
[2] Granroth, G.E., *et al*., Phys. Rev. Lett. **77**, 1616 (1996).
[3] Birk, T., *et al*., Inorg. Chem. **50**, 5312 (2011).
[4] Podlesnyak, A., *et al*., in progress (2012).

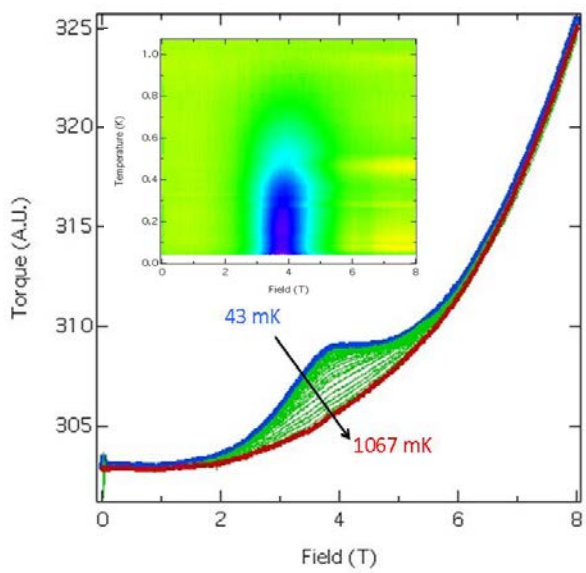

Figure 1. The temperature and magnetic field response of the torque magnetometer, when the external field is applied nominally perpendicular to the chains in a single crystal. (inset) The weak (green) to strong (blue) response (as a color gradient) indicates the presence of a critical field, H_C = 3.8 T.