High Field Magnetic Properties of the Frustrated Cluster Magnet $LiZn_2Mo_3O_8$

J.P. Sheckelton, T.M. McQueen (Johns Hopkins University, Chemistry/Institute for Quantum Matter, Physics and Astronomy); R.D. McDonald, O.E. Ayala-Valenzuela (NHMFL-PFF)

Introduction

Understanding emergent phenomena, where the collective interactions of many simple ingredients gives rise to complex behavior, has led to a strong interest in geometrically frustrated magnetic systems. Building an extended lattice of magnetic clusters, as opposed to single magnetic ions, provides many advantages in stability and tunability. $LiZn_2Mo_3O_8$ is a material in which the magnetic unit is a small Mo_3O_{13} cluster with 1 unpaired e^- /cluster (**figure A**). The interaction of these S=1/2 clusters has been proposed to lead to the formation of a condensed valence-bond state [1]. High field magnetization measurements on $LiZn_2Mo_3O_8$ have been used to probe the nature magnetic interactions on and between Mo_3O_{13} clusters to elucidate the magnetic ground state.

Experimental

Magnetization of $LiZn_2Mo_3O_8$ powder samples was measured on the 65 T short pulse capacitor bank-driven magnet at the NHMFL-PFF with a compensated-coil susceptometer. The data were integrated to extract $M(\mu_o H)$, and scaled to laboratory isothermal magnetization measurements. Electron spin resonance (ESR/EPR) on $LiZn_2Mo_3O_8$ was measured down to T = 1.7 K in the frequency range of 11 GHz to 27 GHz and applied field up to $\mu_o H$ = 2 T using powder samples in an appropriately tuned microwave cavity.

Results and Discussion

The ESR data at T = 1.7 K show a single resonance for $LiZn_2Mo_3O_8$ (**figure, B**). The estimated density of spins giving rise to the ESR signal is in agreement with that expected for one unpaired electron per Mo_3O_{13} cluster. Further, the g-factor extracted from these measurements, 1.91, is very close to the free ion value value of 2.0, suggesting that simple single-ion effects (e.g. spin-orbit coupling) are not the dominant contributor to the low temperature magnetic properties in $LiZn_2Mo_3O_8$. High field magnetization measurements were used to probe the nature of the low-temperature magnetic state. At T = 1.3 K, the magnetization appears to approach a saturation value, that is not explained by Brillouin-type paramagnetic behavior (**figure, C**), even when the symmetry-allowed anisotropic electron g-factor is included. This provides further support for the collective nature of the low temperature magnetic ground state in $LiZn_2Mo_3O_8$.

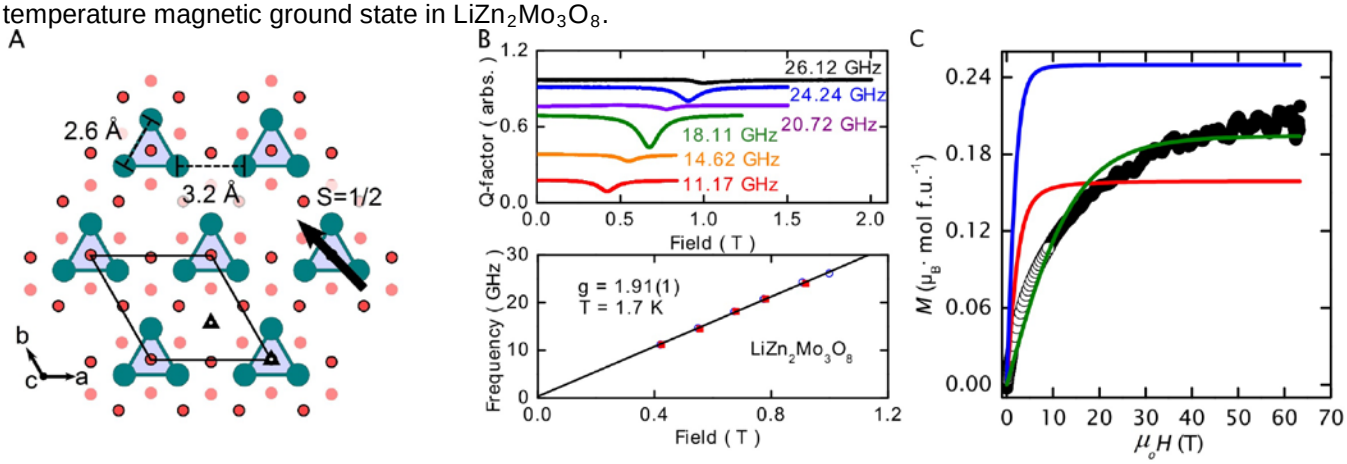

Figure: A, Structure of the $LiZn_2Mo_3O_8$ triangular layer displaying inter- and intra- cluster bond lengths. **B,** ESR showing a single resonance at a g-factor of 1.91. **C**, High field magnetization of $LiZn_2Mo_3O_8$ (Lab PPMS in open circles). The blue line has g_1=g_2=1.9 and g_3=0 (2-D constrained Heisenberg), the red line has g_1=g_2=0 and g_3=1.9 (Ising like), both are scaled for the magnetization of 1/3 of spins per mol f.u. The green line has g_1=g_2=0.3 and g_3=0.1. None of these models adequately describe the observed data, and are inconsistent with the ESR measurements.

Conclusions

We have shown that the magnetic properties of S=1/2 Mo_3O_{13} clusters in $LiZn_2Mo_3O_8$ is not solely due to single-ion physics (*e.g.,* spin-orbit coupling), consistent with a high degree of magnetic frustration in this system.

Acknowledgements

Research under the Institute for Quantum Matter is supported by the US Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award DE-FG02-08ER46544. Work at the NHMFL-PFF was supported by the NSF, US-DOE and the State of Florida.

References

[1] Sheckelton, J.P., *et al.*, Nature Materials, **11**, 493-496 (2012).