Experimental Report

Proposal: **5-42-296** **Council:** 10/2011

Title: Spin and charge correlations in the unconventional spin glass Y2Mo2O7

This proposal is resubmission of: 5-53-215

Researh Area: Physics

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Samples: Y2Mo2O7

Instrument	Req. Days	All. Days	From	To
D7	10	7	26/02/2013	05/03/2013

Abstract:

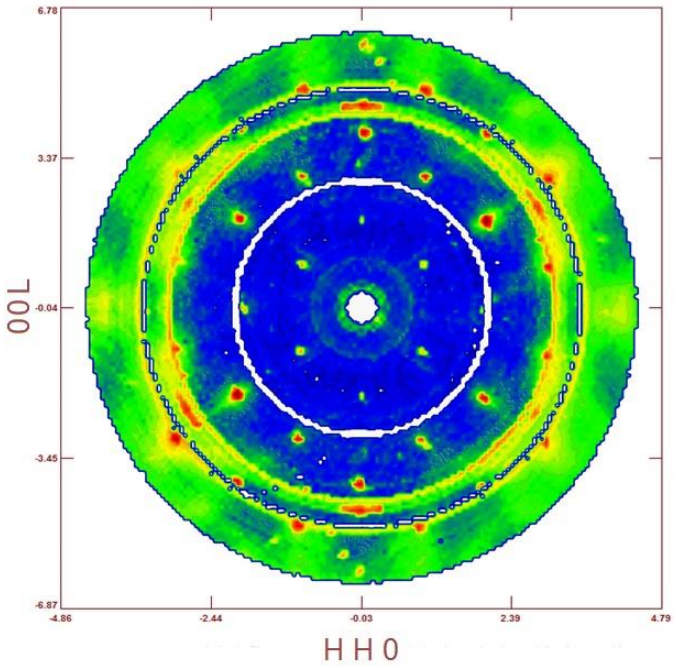
One of the more remarkable aspects of geometrically frustrated magnets is the emergence of spin glass states in the absence of significant amounts of chemical disorder. Conventional solid state theory predicts that frustration and disorder are required for such states, but there have been an increasing number of systems which seem to show only short-ranged ordering at low temperatures with a freezing of magnetic moments. Y2Mo2O7, a pyrochlore lattice with S=1 Mo4+ spins, is perhaps one of the more celebrated examples of these kinds of materials, with an apparent spin-glass transition at T = 22 K. Previous experiments have shown that there is a significant amount of diffuse scattering appearing at high temperatures due to the short-ranged ordering of Mo spins in powder samples. However, until now, there have been no studies on single crystalline samples. Through a collaboration with FSU, UWinnipeg, and NIST, we have grown high quality single crystals of Y2Mo2O7, and have shown through recent NIST experiments on the DCS that significant diffuse scattering is present. With this ILL proposal, we hereby ask for time on D7 to distinguish between charge and spin correlations.

$Y_2Mo_2O_7$ is a pyrochlore oxide that has been the subject of much controversy over the past three decades. Although the material behaves much like a typical spin glass, $Y_2Mo_2O_7$ shows no signs of long-range disorder within the resolution of the probes used to measure it, challenging the foundations of spin glass theory. In 1999, a peak observed at Q=0.44 $Å^{-1}$ on powder $Y_2Mo_2O_7$ using the SPINS triple-axis instrument (NCNR, Gaithersburg, MD) was reported [1]. Over the last 15 years there have been numerous works that indicate a significant amount of bond disorder existing within powder samples. Although theoretical studies suggesting that a spin glass state can result from such bond disorder exist, the energy scale of the relevant experimentally observed interactions suggest that an alternative mechanism might be at play. We have grown a large single crystal of $Y_2Mo_2O_7$ and have used the D7 to investigate the Q-dependence of feature reported earlier in powders [1, 2].

Previous experiments on single crystal $Y_2Mo_2O_7$ have yielded intriguing results. Experiments were performed on the Disc Chopper Spectrometer (NCNR, Gaithersburg, MD) and the 4-ID-D X-ray beamline (APS, Argonne, IL) [2]. Elastic diffuse scattering was observed at Q=0.44 $Å^{-1}$, but remarkably, it appears completely isotropic in reciprocal space (a ring) [2]. Attempts to model the ring using a computationally exhaustive analysis with an isotropic spin Hamiltonian have failed with the constraint that the system is to have the experimentally derived Curie-Weiss temperature θ=-200 K. The diffuse scattering pattern could be successfully reproduced when the dominant interactions are ferromagnetic [2]. Diffuse scattering was also observed at the {222} peaks using both neutrons and X-rays, but the ring could not be kinematically reached using the latter. This left open the question as to whether or not charge correlations were in fact responsible for the spin glassiness.

We used the D7 polarized neutron spectrometer to separate out the diffuse scattering at Q=0.44 $Å^{-1}$. A 3cm single crystal of $Y_2Mo_2O_7$ was oriented on an Al mount in the HHL plane. The orientation was checked and adjusted using OrientExpress. Incident wavelengths of 3.12 Å and 4.855 Å were used, the latter corresponding to a reciprocal space coverage of 0.09 $Å^{-1} < Q < 2.5$ $Å^{-1}$. Data was collected over 3 days at each wavelength below T_g. The entirety of the ring was observed in the spin flip channel below the glassy temperature T_g = 22.5 K, while only a portion of the diffuse scattering surrounding the {222} was observed in the spin flip channel (Figure 1). This unambiguously demonstrates the magnetic character of the ring. A quantitative analysis is currently underway.

[1] – Gardner *et al.* (1999) Phys. Rev. Lett. **83**, 211-214.
[2] – Silverstein *et al.* (2014) Phys. Rev. B, *in press*.



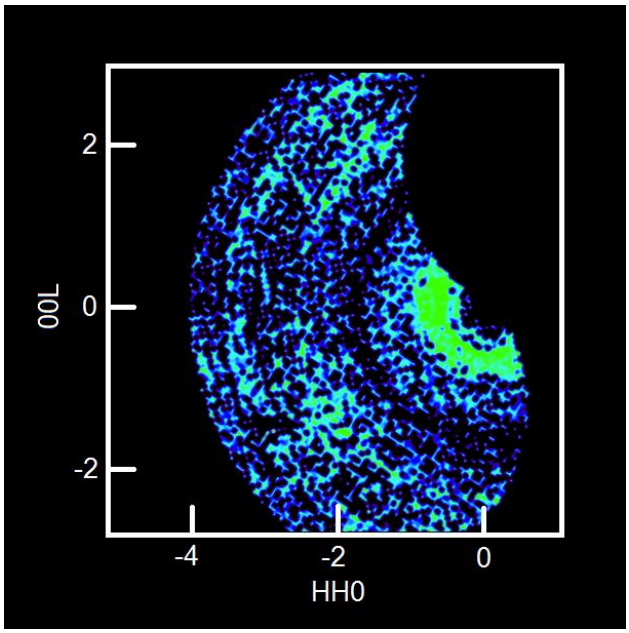


Figure 1: Top) Raw data from LAMP of the total scattering using λ = 3.12 Å above T_g; **Bottom)** The spin flip channel at T = 2 K using a wavelength of λ = 4.855 Å. The data is folded over one quadrant to improve the low statistics and rotated into the correct orientation. A clear ring is visible surrounding the origin as well as surrounding the (22-2) and (-2-2-2) reflections, consistent with the earlier findings in [2]. This unambiguously demonstrates the magnetic character of the diffuse scattering.