Experimental report

05/02/2018

Proposal: **5-42-449** **Council:** 10/2016

Title: Supercriticalilty and the Widom Line in Holmium Titanate

Research area: Physics

This proposal is a new proposal

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Samples: $Ho_2Ti_2O_7$

Instrument	Requested days	Allocated days	From	To
D7	7	7	28/02/2017	07/03/2017
IN3	2	2	09/02/2017	11/02/2017

Abstract:

Supercritical phenomena in conventional fluids remains a difficult subject to study experimentally, requiring high temperatures and pressures to realize. An experimental candidate for study of supercritical physics reveals itself in the classical spin ices $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$, whose well known ground state behavior and zero point entropy is a measurable analogue of diamond lattice water ice. Recent susceptibility measurements of the spin ice $Dy_2Ti_2O_7$ suggest that the magnetic Coulomb gas of emergent monopoles has an analogous crossover in thermophysical properties in the supercritical region above the well-known liquid-gas critical point, demonstrably similar to the Widom line in conventional supercritical fluids. We propose to study this effect in $Ho_2Ti_2O_7$, by measuring the evolution of the diffuse scattering across the Widom line. By comparison with susceptibility experiments, and Monte Carlo simulations of both Coulomb gases and spin systems, we aim to achieve a full microscopic understanding of the Widom line.

Supercriticalilty and the Widom Line in $Ho_2Ti_2O_7$

Proposal No.: 5–42–449

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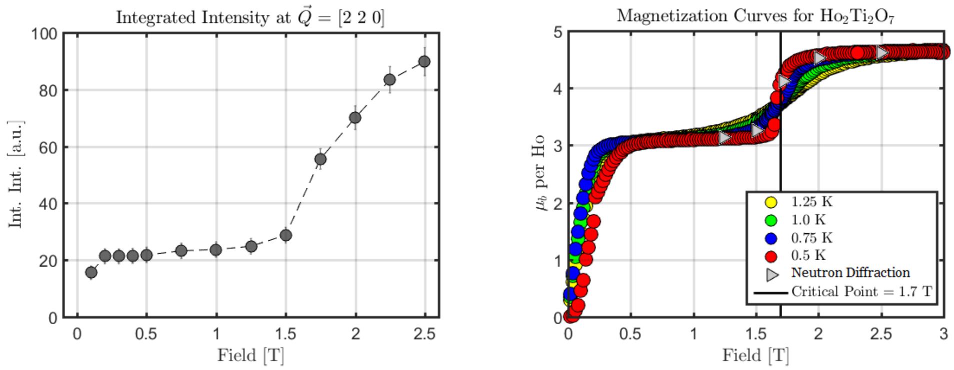
Instrument: D7 **Date:** 27.02.17 – 7.03.17
Instrument Responsible: Andrew Wildes

Abstract: Diffraction measurements with the uniaxial polarization analysis setup on D7 were made in the $[\bar{1}\ 1\ 0]$–$[\bar{1}\ \bar{1}\ 2]$ plane of a series of single crystal samples of $Ho_2Ti_2O_7$ aligned in a copper holder for perpendicular fields between 0 and 2.5 T at 0.7 K to investigate the supercritical regime of the kagomé ice phase of $Ho_2Ti_2O_7$.

Introduction: In the geometrically frustrated pyrocholres $Ho_2Ti_2O_7$ and $Dy_2Ti_2O_7$, the large magnetic moments are constrained to point along the easy axis of the corner sharing tetrahedra. The resultant 2-in/2-out energy minimizing configuration hosts a magnetic Coulomb phase with characteristic pinch points [1,2]. Applying a field along the $\langle 111 \rangle$ direction of these spin ices pins the one spin per tetrahedra aligned along that axis. The remaining three spins in the kagomé plane can fulfill the ice rules in a phase known as kagomé ice [3,4]. Changes in the density of single spin-flip excitations from the kagomé ice ground state, recontextualized as magnetic monopole quasi-particles [5,6], could potentially manifest as a peak in the adiabatic susceptibility reminiscent of a Widom line in a supercritical fluid [7-9].

Experimental Methods: Previous experiments have observed the effect that a tilted field [10] and demagnetization [11] has on reducing the diffuse scattering structure factor symmetry. To eliminate influence from a strong shape-dependent demagnetization effect while maintaing a robust size, a sample of single crystal $Ho_2Ti_2O_7$ was grown at PSI, aligned, and cut into prisms of with dimensions 2 x 2 x 8 mm. A copper holder was designed to align the samples in parallel, and the samples were aligned on OrientExpress and IN3 preceding the experiments, and a singular sample was used in the final measurements. The diffuse scattering patterns still reveal traces of a tilted field, and will be analyzed as such. Background estimates were collected with no sample, vanadium, cadmium, and quartz samples (21 repetitions, 2:1 spin flip/non-spin flip) with transmission values from self attenuation measurements of 12769, 7795, 71, 12678, and 10140 respectively. 180° scans were taken on D7 at $2\theta = 75$ and 79.5°, with a wavelength of 4.855 Å to optimize accessible $\vec{Q}$, flux, and resolution.

Results: The magnetic bragg peak at $\vec{Q} = [2\ 2\ 0]$ was measured with a variety of fields to confirm the presence of the magnetization plateau in $Ho_2Ti_2O_7$.



(a) (b)

Figure 1: (a) Measured integrated intensities of the bragg peak at $\vec{Q} = [2\ 2\ 0]$, 0.7 K with field, demonstrating the expected magnetization plateau with a field along the [1 1 1] axis; (b) Magnetization measurements for $Ho_2Ti_2O_7$ plotted with field points for the D7 omega scans

Diffraction patterns were then taken at 1.25, 1.5, 1.75, 2, and 2.5 Tesla at 0.7 K and the change in intensity and location of peaks and pinch points in the structure factors across the critical point were observed. The uniaxial polarization analysis setup on D7 was used to separate the behavior of spin components in the kagomé plane (as measured in the spin flip channel) from the Ising antiferromagnetic spin components parallel to the applied field (as measured in the non-spin flip channel).

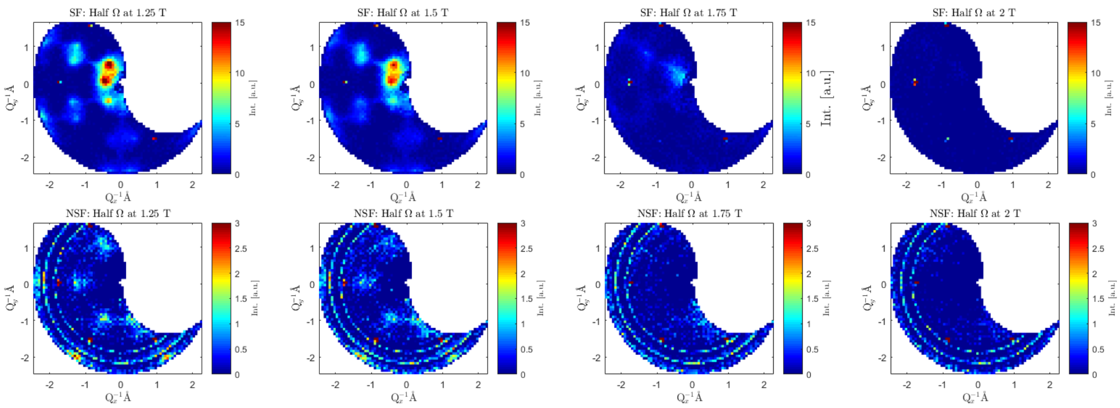


Figure 2: A series of half Ω scans in the SF channel (first row) and NSF channel (second row, visible copper powder lines) at 1.25, 1.5, 1.75, and 2 T with a subtracted background

The pinch point widths in the spin flip channel at $\vec{Q} = (-0.88, \pm 0.5)$ Å^{-1} were fitted with a lorentzian from the diffraction patterns measured in the spin flip channel and rotated to capture the full longitudinal widths. These widths were determined and plotted with field above the critical temperature for $Ho_2Ti_2O_7$.

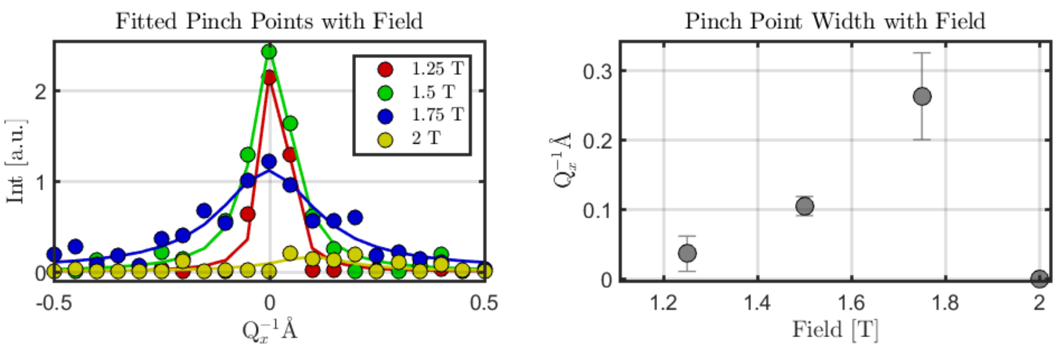


Figure 3: Refined intensities fitted and widths plotted against field

Despite the difficulty in creating a uniformly magnetized sample, our experiment provided evidence for the presence of short range spin correlations at supercritical temperatures and a decrease in correlation length with increasing field. High frequency AC susceptibility measurements have been taken at Cardiff University, and corroborate the presence of a peak in the adiabatic susceptibilty in $Ho_2Ti_2O_7$ at the expect location of the Widom line.

References:

[1] Bramwell & Gingras, Science **294**, 5546 (2001); [2] Fennell *et al.*, Nat. Phys. **3**, 8 (2007); [3] Moessner & Sondhi, Phys. Rev. B **68**, (2003); [4] Harman-Clarke, UCL (University College London), 2012); [5] Castelenovo *et al.*, Nature **451**, 7174 (2008); [6] Higashinaka *et al.*, PRL **95**, 23 (2005); [7] Bovo *et al.*, Nat. Comm. **4**, 1535 (2013); [8] Simeoni *et al.*, Nat. Phys. **6**, 7 (2010); [9] Krey *et al.*, Phys. Rev. Lett. **108**, 257204 (2012). [10] AT & TF, unpublished. [11] Twengström *et al.*, Phys. Rev. Materials **1** 044406 (2017);