Experimental report

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Proposal: 5-32-856 **Council:** 4/2018

Title: Novel phases in spin ice accessed via the application of pressure.

Research area: Physics

This proposal is a new proposal

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Samples: Dy2Ti2O7

Instrument	Requested days	Allocated days	From	To
D7	5	5	09/10/2018	14/10/2018

Abstract:

The most prominent members of the spin-ice group are Ho2Ti2O7 (HTO) and Dy2Ti2O7 (DTO), in which magnetism arises from localised rare-earth ions with a strong crystal-field anisotropy. These exchange interactions lead to the exotic behaviours which, in the case of spin ice materials, includes residual entropy, magnetic monopoles and a Coulomb phase. Neutron scattering has been significant in the determination of these exotic states. Under applied pressure, spin-ice compounds on the pyrochlore lattice are predicted to display exotic magnetic behaviour that included an infinite-order multicritical transition between a Coulomb liquid phase and an ordered magnetised state. Recent susceptibility measurements on DTO reveal strongly enhanced antiferromagnetic correlations upon the application of hydrostatic pressure. This proposal aims to quantify these magnetic correlations using polarisation analysis on the diffuse scattering spectrometer, D7.

Novel phases in spin ice accessed via the application of pressure (5-32-856).
I.M. Bakke, R. Edberg, K. Lefmann, P. P. Deen & A. R. Wildes.
D7 (ILL), 09 - 14 October 2018.

Experimental abstract: The most prominent members of the spin-ice group are Ho2Ti2O7 (HTO) and Dy2Ti2O7 (DTO), in which magnetism arises from localised rare-earth ions with a strong crystal-field anisotropy. These exchange interactions lead to the exotic behaviours which, in the case of spin ice materials, includes residual entropy, magnetic monopoles and a Coulomb phase. Neutron scattering has been significant in the determination of these exotic states. Under applied pressure, spin-ice compounds on the pyrochlore lattice are predicted to display exotic magnetic behaviour that included an infinite-order multicritical transition between a Coulomb liquid phase and an ordered magnetised state[1-5].
The experiment will measure the magnetic correlations in 0.2 g powdered DTO using XYZ PA at ambient pressure at 1.5 K (24 hours) in the pressure cell (but not pressurised), followed by a measurement at 1.2 GPa for which the sample will have to be removed and the pressure applied external to the instrument. A range of temperatures will be measured (20 K, 5 K 2K, and base temperature, T 1.5 K (24 hours each)). Background and polarisation calibrations will require an extra day. A total of 5 days of beam time is requested.

The experimental team were unable to procure $Dy_2Ti_2O_7$ with the correct isotope. Instead the experiment on D7 measured $Ho_2Ti_2O_7$ under uniaxial pressure at 1.5 K. A uniaxial pressure cell developed with the help of the Scientific Activity Division (SAD: M. Guthrie and A. Holmes) of the ESS enabled the application of uniaxial pressure after in-house calibration (L. Sandberg). A single crystal of $Ho_2Ti_2O_7$ was grown within the team (I.M Bakke & L. Sandberg). 5 single crystal $Ho_2Ti_2O_7$ samples were laser cut to provide an axis for the application of uniaxial pressure along the [0 0 1] (3 samples), [1 1 0] (1 sample)and [1 1 1] (1 sample) directions. The sample dimensions were height x diameter: 3 x 2 mm, with a calculated weight of 65 milligrams.

The experimental conditions were:

- Instrument D7, ILL (51.4 MW Reactor power). 09-14 October 2018.

- $\lambda = 4.8621$ Å calibrated using a YIG sample, calibration 19th September 2018.
- Sample 1 [0 0 1], scattering plane (h 0 0) & (0 k 0), access via a rotation of the sample (180 degrees). For interest: D7 Q resolution function: Nuc.Instr.&Meth. Pays. A 857 24-30 (2017). Positioned with zero, 2 and 4 kN applied force at room temperature. Calibration estimates that an extra 3 kN is applied at 1.5 K due to cell contraction. This indicates that the forces applied are 3, 5 and 7 kN providing uniaxial pressures of 0.95, 1.59 and 2.22 GPa respectively on the sample at 1.5 K.
- Beam depolarised for the x and y directions - only slightly for the z direction. Flipping ratios (on Bragg peak): Z $\sim$ 16, X $\sim$ 3, Y $\sim$ 2.5. Suspect depolarisation due to the stainless steel anvils.
- Slit width and height before the sample were optimised on a Vanadium sample 3 mm high & 8 mm wide.
- Pressure cell was covered in Cd to minimise spurious scattering, see Figure below.
- The Q resolution of D7 was not sufficient to determine any changes in lattice parameter.
- A background measurement of the cell was performed using an Aluminium nut at the sample position (to ensure the anvils did not close)
- Detector and polarisation calibrations from a previous experiment were used due to time limitation. It was estimated that a good estimate of the absorption was more important.
- The magnetic signal is obtained in the spin flip channel with Z polarisation analysis. The scattering therefore contains some incoherent contributions which is small in the case of HTO.
- The absorption of the sample, as a function of rotation angle has been obtained using the, nominal, paramagnetic signal of the sample under pressure (4 kN at RT) at 50 K.
- To increase the pressure the sample was brought up to room temperature and further force was applied using a calibrated force press. Sample 1 was pressurised up to 0.95 GPa and removed. Sample 2 (also [0 0 1]) was pressurised up to 1.59 and 2.22 GPa without removal.

In the following figures the raw data is presented. Raw data is considered data that has been corrected for detector and polarisation efficiency but not corrected for absorption. The (4 0 0), (0 4 0) and variations on (2 2 0) were determined (not shown). Figure 1 (top,left) shows the magnetic scattering for sample 1 at 0.95 GPa (0 kN applied at RT). Figure 1 (top,right) shows the magnetic scattering for sample 2 at 1.59 GPa (2 kN applied at RT).

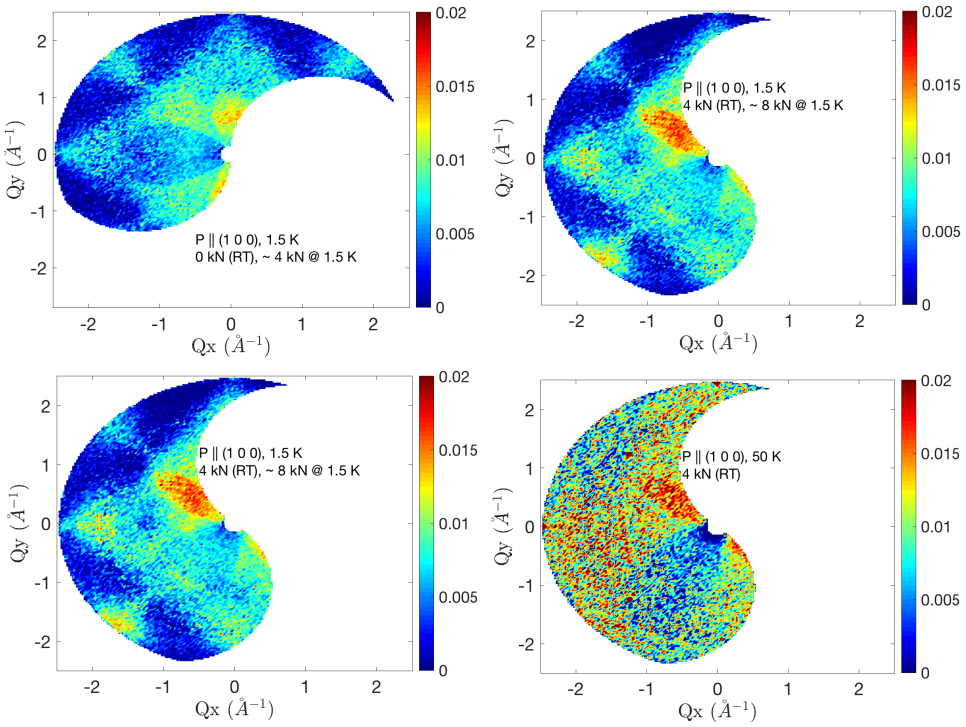

Figure 1: Raw data, Spin flip scattering with Z polarisation for (top, left) 0.95 GPa, (top, right) 1.59 GPa, (bottom, left) 2.22 GPa and (bottom, right) Paramagnetic scattering.

Figure 1 (bottom,left) shows the magnetic scattering for sample 2 at 2.22 GPa (4 kN applied at RT). Figure 1 (bottom,right shows the magnetic scattering for sample 2 at 2.22 GPa (4 kN applied at RT) at 50 K which should provide paramagnetic scattering. In order to correct for the absorption of the cell one can assume that the paramagnetic scattering is equal in a detector for the complete rotation of the crystal. As such it should be possible to quantify the scattering losses as a function of detector angle and rotation and normalise the scattering data by this. Data analysis is progressing.