Experimental report

13/09/2018

Proposal: **5-31-2545** **Council:** 4/2017

Title: Effect of pressure on the magnetism of the two-dimensional antiferromagnet FePS3

Research area: Physics

This proposal is a new proposal

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Samples: FePS3

Instrument	Requested days	Allocated days	From	To
D20	5	5	05/09/2018	10/09/2018

Abstract:

We have metallised the two-dimensional insulating honeycomb antiferromagnet FePS3 at 5.5 GPa. The mechanisms for metallisation, as well as the physical properties of the metallic state, are not known. We wish to determine the magnetic and structural phase diagram as a function of pressure. This has never been measured and will offer valuable insight into this material.

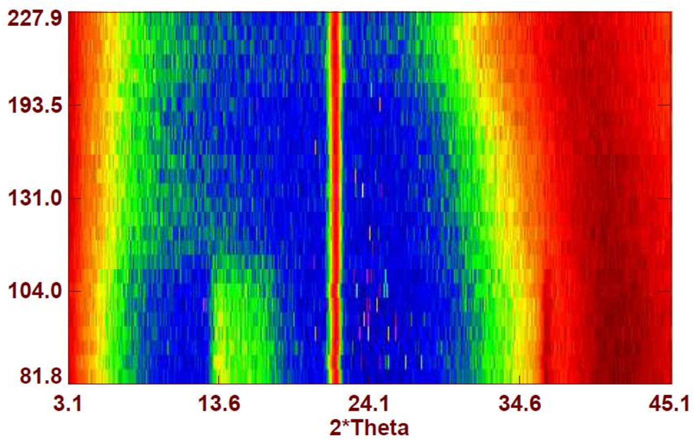
Exp 5-31-2545 High pressure powder diffraction on $FePS_3$ on D20

Experimental report

Our goal in this experiment was to observe the evolution of the magnetism and magnetic structure with temperature and pressure in 2D Ising antiferromagnet $FePS_3$. Our previous measurements of electrical transport and of the crystal structure via synchrotron radiation have shown multiple structural transitions and a Mott insulator - metal transition at high pressure; we wish to study how the magnetism evolves as we tune the system from 2D to 3D and from isolated electronic sites to metallic state.

We used monochromatic neutrons with wavelength 2.42 Angstroms and a single-toroidal Paris-Edinburgh type pressure cell with cadmium-clad boron nitride anvils and titanium-zirconium gasket. Methanol-ethanol was used as pressure medium and care was taken to keep the temperature sufficient to maintain the pressure medium in its liquid state when changing pressure. Lead, and the known equation of state of its lattice parameters, was used as a manometer.

Initially, we measured the diffraction patterns upon both cooling and warming through the Néel Temperature to check for the emergence of any thermal hysteresis in the transition, but with ~30 minutes (and at higher pressures, up to 60 minutes) required to acquire each scan and the limited time available, we were only able to realistically achieve temperature resolutions of around 4 K. After the first 2 pressure points we instead adopted a regimen of cooling rapidly with the use of liquid nitrogen from room temperature and acquiring data while continuously warming. The large thermal mass of the PE press demands a large input of heater power, and a concern is that thermal gradients within the setup may lead to uncertainty of the true sample temperature, so we attempted to keep warming profiles systematic.



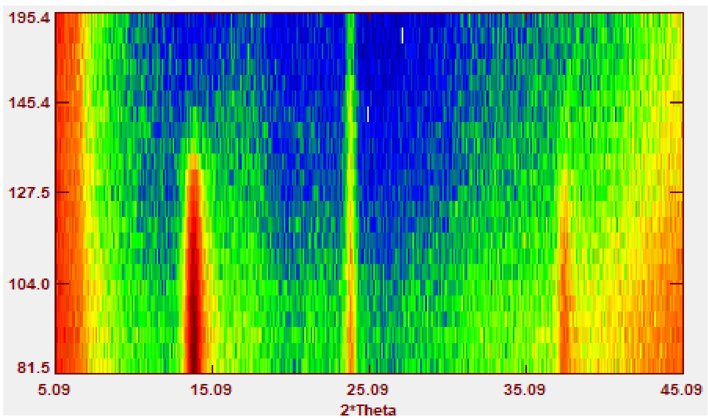


Fig. 1 Normalised colour plots of the neutron diffraction patterns of FePS3 as a function of temperature (vertical axis, in K). Upper plot is at ambient pressure, lower at 7 GPa. Characteristic magnetic Bragg peaks were observed as with previous ambient pressure measurements at around 13 and 35 degrees 2ϴ in the antiferromagnetic phase.

A thorough analysis is required, but the Neel temperature (118 K at ambient) can be seen to continuously increase across such plots as pressure was raised, reaching 145 K at 7 GPa. The red region to the right of the top plot is the diffuse scattering of the pressure medium.

The temperature was raised to room temperature on each pressure run, both to melt the pressure medium and to allow us to match the structural peaks seen with our previous synchrotron high-pressure data. A base temperature of 80 K was used, as this was easily and quickly achievable and well within the antiferromagnetic phase of the sample. We observed a drop in overall signal by up to 50% while warming from base to room temperature. This was particularly pronounced at the highest pressures due to the small separation of the anvils at this point. This is a known issue, but with only speculation as to the cause (tilting of the cell due to uneven thermal contraction?) on D20. Our interest was focussed below 150 K so this was not too impactful to our data, but careful normalisation of the datasets will be required to account for this effect.

From our previous measurements we know of a structural distortion of the unit cell at pressures between 3-5 GPa in $FePS_3$, and a collapse of the interlayer spacing accompanying the Mott insulator-metal transition at around 14 GPa. With the setup used on D20 we were able to access approximately 8 GPa (the signal of the lead manometer was poor at the highest pressure and a formal analysis will be required to estimate the final pressure). This was sufficient range to observe the magnetic evolution over a wide range of the phase diagram, and we hope to gain information about the effect of the first structural distortion on the magnetic structure from the data, but a more specialised setup will be needed for future experiments to reach the metallic phase.

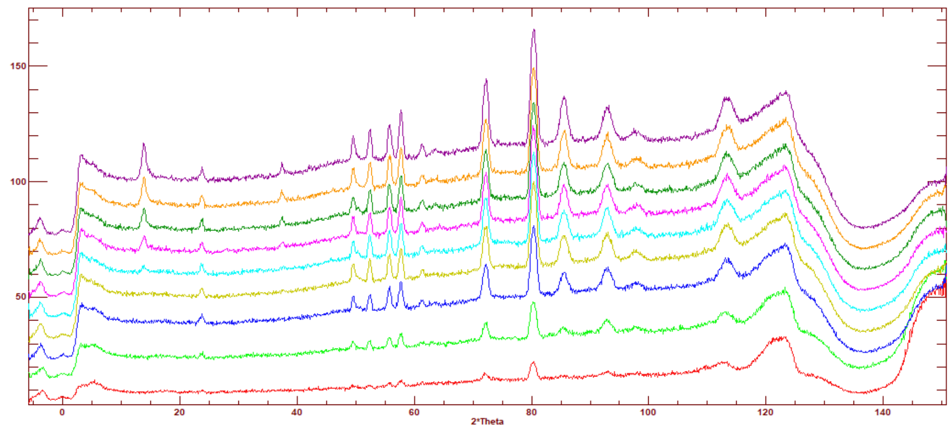


Fig. 2 Diffraction patterns taken while warming the sample at 7 GPa. Topmost pattern is at 80 K, bottom is at room temperature. A subset of curves is shown and they are offset for clarity.

Example individual patterns are shown in Fig. 2. Structural Bragg peaks were clearly resolved, and can be matched onto our high-pressure x-ray data, and magnetic peaks can be seen to emerge below the magnetic ordering temperature. to either side of the structural 001 peak at ~23 degrees. Interestingly, the lowest-angle magnetic peak was seen to sharpen into a single feature, rather than the extended form seen at ambient pressure due to 2D rod-like correlations - perhaps an indicator of changes in dimensionality. This can be crudely observed in the maps shown in Fig. 1. The Neel Temperature additionally shifted continuously upward as magnetic exchange between the planes was strengthened by the application of pressure, in agreement with, but greatly extending, preliminary susceptibility measurements up to 1 GPa. This experiment proved the first direct measurement of the magnetic structure in this wide family of materials under hydrostatic pressure.