Experimental report

15/02/2023

Proposal: 4-01-1751 **Council:** 4/2021

Title: Magnetic dynamics in the high-pressure phase of antiferromagnet FePS3

Research area: Physics

This proposal is a new proposal

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Samples: FePS3

Instrument	Requested days	Allocated days	From	To
PANTHER	4	4	01/07/2021	05/07/2021

Abstract:

The MPX3 materials (M = Fe, Ni, Mn; X = S, Se) are a family of quasi-2D antiferromagnetic insulators, with most sharing a similar monoclinic unit cell and C2/m space group. The metal M2+ ions form a honeycomb lattice in the ab planes, which are weakly coupled by van der Waals forces. These layered magnets are of significant current interest in low-dimensional materials communities.

Our previous studies show that FePS3 undergoes two structural transitions under pressure, the second of which coincides with the metallisation of the system. Through record breaking high-pressure neutron diffraction at the ILL we have shown that both of these transitions induce changes in the magnetic structure of FePS3, by changing the inter-layer coupling and introducing a new form of short-range magnetic order at the highest pressures.

We propose here to develop this further by measuring the evolution of magnetic dynamics into the first high pressure phase. We will use high-pressure apparatus at low-temperature on PANTHER to measure inelastic scattering above 4 GPa. This will develop our understanding of these materials whilst also advancing the capability for high-pressure experiments at the ILL.

Experimental report for

4-01-1751: Magnetic dynamics in the high-pressure phase of antiferromagnet $FePS_3$

1 – 6 July 2021

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The experiment aimed to measure the magnetic density of states for $FePS_3$ as a function of applied pressure.

$FePS_3$ is a layered van der Waals antiferromagnetic insulator. Its crystal structure at ambient pressure has a monoclinic space group $C\,2/m$ with angle $\beta \sim 107.2°$, and its magnetic structure below the Néel temperature of $T_N \sim 125$ K has a propagation vector of $\mathbf{k}_M = [01½]$ [RBrec]. The spin waves in the ground state have spectral weight between 15 and 40 meV [AWil]. On applying a hydrostatic pressure, the compound undergoes a structural phase transition at P ~ 4 GPa involving a shear of the layered *ab* planes along the crystallographic *a* axis such that the space group $C\,2/m$ is retained but β becomes almost 90° [CHaines,DJarvis]. The structural transition is accompanied by a change in the propagation vector to $\mathbf{k}_M = [010]$ [MCoak]. A second transition to the higher symmetry space group $P\bar{3}1m$ occurs at P ~ 14 GPa, concomitant with an insulator-metal transition [CHaines]. The magnetism is not quenched in the metallic phase, but the long-ranged order is replaced by short-ranged correlations [MCoak].

The experiment aimed to measure the evolution of the magnetic fluctuations in powdered $FePS_3$ through the first transition at P ~ 4 GPa. PANTHER is the appropriate instrument for such an experiment given the dynamic range of the magnetic spectral weight. The VX-5 Paris-Edinburgh cell with its CCR for low-temperature control was used as the sample environment. The cell was equipped with ceramic Al_2O_3 anvils for greater transparency, TiZr gaskets were used to reduce signal contamination from the sample environment, and the powdered sample was loaded together with a methanol-ethanol mix as a pressure medium. No pressure indicator (e.g. Pb) was included with the sample.

The first day was spent aligning the instrument and preparing the PE cell. The sample was then loaded into the cell. It was initially subjected to an estimated 2 GPa and was cooled to 80 K before measuring for ~ 24 hours. The cell was then warmed to 240 K and the pressure was increased to an estimated 6 GPa before cooling again to 80 K. Another measurement of ~ 24 hours was performed. The cell was then warmed, the sample was removed, and a measurement of the cell containing an empty gasket was performed for ~ 12 hours to estimate the instrument background.

Some technical issues concerning the heating of the cell occurred during the experiment which slowed considerably its warming, reducing the number of pressures that could be measured.

However, the biggest issue was that no obvious scattering from $FePS_3$ could be observed. This could only be properly ascertained once the data were corrected for the background from the PE cell, taken from the measurements for the empty cell performed at the end of the experiment. Figure 1 shows the data at 80 K and nominal P = 2 GPa from a fixed energy window around the elastic line. The simulated scattering from $FePS_3$ and from Al_2O_3, used for the anvils, at ambient pressure and temperature are also indicated. Diffraction from FePS3, measured under similar conditions on D20, is shown in Figure 2. Bragg peaks from the Al_2O_3 are clearly seen. However,

aside from a possible feature at Q ~ 2.5 $Å^{-1}$ that is also a Bragg peak position for Al_2O_3, no clear peaks from $FePS_3$ may be observed.

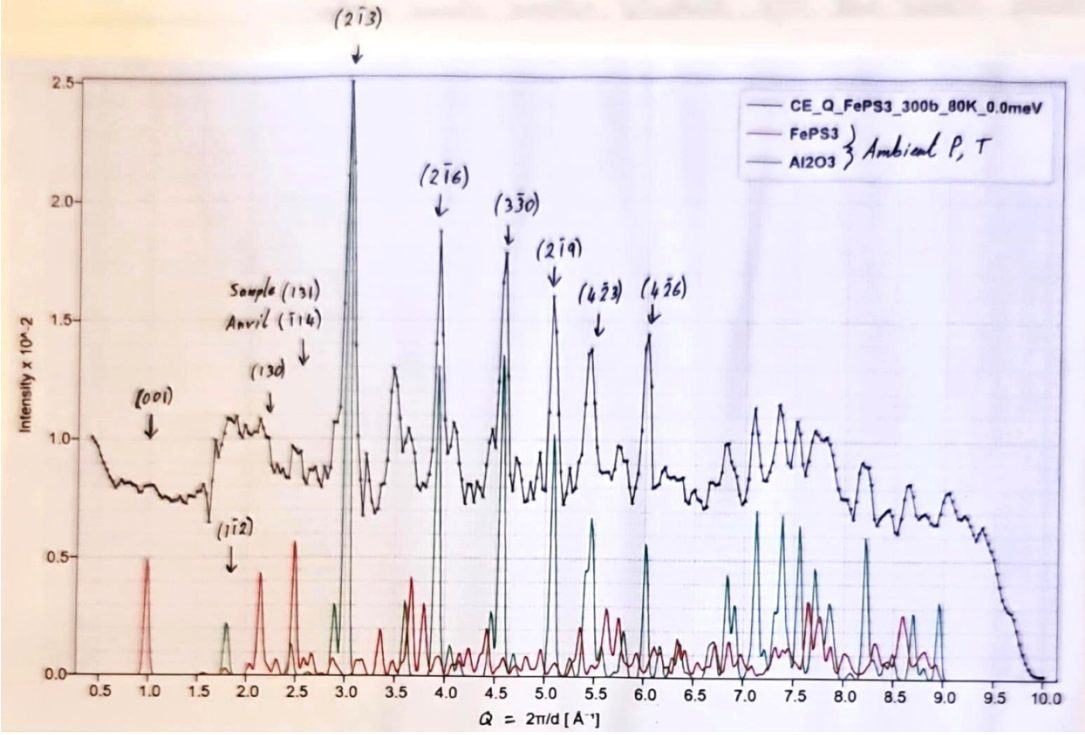

Figure 1: Measured scattering within a fixed energy window around the elastic line from $FePS_3$ in the PE cell at 80 K and an applied nominal pressure of 2 GPa. The simulated diffraction patterns for $FePS_3$ and Al_2O_3 at ambient pressure and temperature are shown as a red and line respectively.

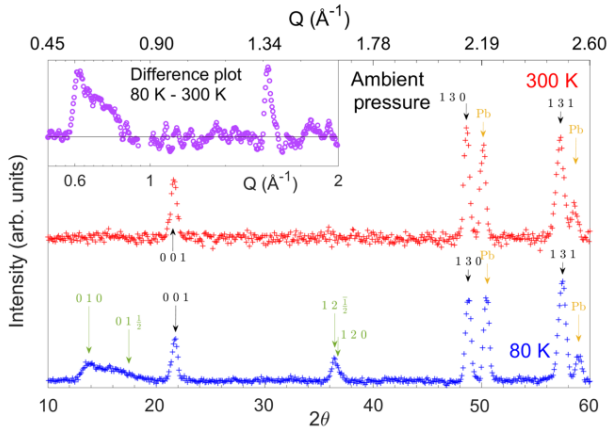

Figure 2: The neutron powder diffraction for $FePS_3$, measured on D20 in the double-toroid Paris-Edinburgh pressure cell at a low applied pressure [MCoak].

A subsequent experiment using the same pressure cell on IN5 to measure the pressure-dependence of the spin wave spectra in the sister compound $MnPS_3$ was highly successful, with clear Bragg peaks from the sample and observable spin wave scattering [ExpRepIN5]. $FePS_3$ with S = 2 has a magnetic moment only slightly smaller than that for $MnPS_3$ with S = 5/2, hence the experience on IN5 suggests that the $FePS_3$ experiment on PANTHER should be feasible. The failure of the experiment to observe any scattering from the $FePS_3$ suggests that there may have been an issue with the alignment of the PE cell such that the sample was masked by the anvils. If careful tests of the cell alignment can be performed and expected scattering intensity verified, using a strong and unambiguous scattering standard like nickel in the sample cavity, it would be worthwhile to make another attempt to perform the experiment.

References:

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[MCoak] M. J. Coak et al., Phys. Rev. X **11** (2021) 011024.
[ExpRepIN5] ILL Experimental Report 4-01-1728