# Experimental report

13/09/2019

**Proposal:** **5-31-2646**

**Council:** 10/2018

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

**Research area:** Physics

**This proposal is a continuation of 5-31-2545**


**Main proposer:** **Matthew COAK**

**Experimental team:** Andrew WILDES
Stefan KLOTZ
Hayrullo HAMIDOV
Cheng LIU
Matthew COAK
David JARVIS

**Local contacts:** Thomas HANSEN


**Samples:** FePS3

| Instrument | Requested days | Allocated days | From | To |
|---|---|---|---|---|
| D20 | 7 | 7 | 17/07/2019 | 24/07/2019 |


**Abstract:**

The MPX3 (M = Fe, Ni, Mn and X = S, Se) family are quasi-2D antiferromagnets which crystallise with a monoclinic unit cell and a space group of C2/m. The transition metals form a very nearly perfect honeycomb lattice in the ab planes. The planes are bound by van-der-Waals forces and the compounds are enjoying a huge surge of interest from the graphene and low-dimensional materials community with recent studies demonstrating the persistence of magnetic order down to single atomic layers.

Our goal in this experiment is to observe the evolution of the magnetism and magnetic structure with temperature and pressure in FePS3. 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 a metallic state.

We have just completed a series of measurements on D20 from 0-8 GPa and 80-300 K using the standard Paris-Edinburgh press - we propose to extend these measurements to higher pressures using a double-toroidal pressure cell.

# Experimental Report - 5-31-2426, continuation of 5-31-2425

## Effect of pressure on the magnetism of the two-dimensional antiferromagnet $FePS_3$

M.Coak, Sept 2019

As in the previous experiment 5-31-2426, we aimed to investigate the evolution with pressure of the magnetic structure of the two-dimensional antiferromagnet $FePS_3$. Our previous experiments using synchrotron x-rays have shown two structural transitions with pressure in this material, the first a sliding transition of the honeycomb layers at ~4 GPa (preserving the C2/m space group) and a second involving a collapse of the interlayer spacing at ~14 GPa (to a trigonal P-31m structure). We have also observed a Mott insulator to metal transition at a pressure coinciding with the second structural transition (Phys. Rev. Lett. **121**, 266801). During our previous experiment on D20 it was only possible to achieve a maximum pressure of 8 GPa, and so it was not possible to measure the highest pressure metallic phase and to discern what changes to the magnetism occur with metallisation.

To reach this phase in the current experiment a Paris-Edinburgh type pressure cell with double-toroidal sintered diamond anvils was used, with a methanol-ethanol pressure medium. This represents the first use of a cell of this type at the ILL for a user experiment. The cell worked exactly as designed, with usable data up to a maximum achieved 183 kbar (18.3 GPa). Knowledge of the crystal structure and lattice parameters of the sample itself under pressure was used for manometry to minimize any unwanted signals in the data. A wavelength of 2.42 Angstroms was used in the powder diffraction geometry of D20 with two-theta scans to even out detector errors.

A room-temperature measurement at the minimum gas pressure to safely seal the cell was performed first. Subsequently measurements were performed at the pressures shown in Figure 1**Error! Reference source not found.**, to give good coverage of all three structural phases. Sample pressure values will require more formal data analysis to verify these estimates.

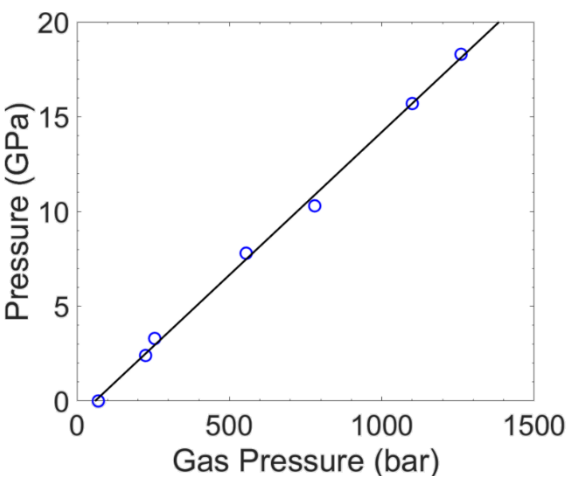


*Figure 1 - Calculated sample pressure vs gas loading pressure for meassurements.*

Measurements at 3.3 and 7.8 GPa were made to coincide with data from the previous experiment, with scans being made at 300 K and a base temperature of 80 K - below the magnetic transition (118 K at ambient and increasing with pressure). Measurement of the Neel temperature and some evolution of the magnetic order at each pressure was performed by collecting data whilst warming from 80 to 300 K, though at higher pressures the necessarily longer count times rendered this imprecise to the order of 10 K.

A strong peak was seen in the data from the sintered diamond anvils. Additional peaks which did not move with applied pressure we were able to index as diamond, but with a wavelength of a third of the assumed value - a small percentage of lambda/3 neutrons appear to have passed though the collimator. These peaks are easily seperable from the data as they do not shift with increasing pressure.

We were able to track the evolution of the magnetic structure through Bragg peaks present at low but not high temperature. We could also measure the Neel temperature from the integrated intensity of these peaks. A key result which now requires theoretical input and significant data analysis is the appearance of a broad magnetic feature at the highest pressures (shown in the below plots). This cannot easily be indexed with trial magnetic propagation vectors and is very broad - suggesting a form of short-range-order. This feature emerges around the metallisation pressure, and accompanies the suppression of the long-range antiferromagnetic order. Intriguingly, it persists, with decreasing magnitude, to temperatures above room temperature.