Experimental report

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Proposal: 5-41-1177 **Council:** 4/2021

Title: The high-field magnetic structure of FePS3

Research area: Physics

This proposal is a new proposal

Main proposer: Andrew WILDES

Experimental team: Andrew WILDES
Jennifer GRAHAM
Fabienne DUC
THIERRY LEMAIRE
Matthew COAK

Local contacts: Andrew WILDES
Frederic BOURDAROT

Samples: FePS3

Instrument	Requested days	Allocated days	From	To
IN22	9	8	05/10/2021	13/10/2021

Abstract:

FePS3 is a quasi-two-dimensional Ising-like antiferromagnet. Two magnetic transitions are observed on applying a magnetic field normal to the planes, which is also the ordered moment direction. The first transition occurs at H~38 Tesla and results in a plateau at half the saturation magnetisation, i.e. M/Msat = 1/2, while the second transition is to the saturation magnetisation and occurs just over 40 Tesla. A M/Msat = 1/2 plateau is in contradiction with the position of FePS3 on the calculated magnetic phase stability diagram. We wish to determine the magnetic structure in the M/Msat = 1/2 plateau to confirm the phase-stability diagram and to help refine the underlying Hamiltonian for FePS3.

Experimental report for 5-41-1177: The high field magnetic structure of $FePS_3$

IN22, 5 – 13 October 2021

Experimental team: Frédéric Bourdarot, Matt Coak, Fabienne Duc, Jennifer Graham and Andrew Wildes

$FePS_3$ is a layered van de Waals compound, forming long-ranged antiferromagnetic order below its Néel temperature of 125 K [1, 2]. The Fe^{2+} moments form a honeycomb structure in the ab planes, and it has a strong Ising-like anisotropy with the moments pointing normal to the planes.

Two phase transitions are observed in magnetization measurements when applying a strong magnetic field along the aligned moment direction [3, 4]. The first occurs at ~39 T from magnetization $M = 0$ to $M/ M_{sat} = ½$, where M_{sat} is the saturation magnetization. The magnetization plateau persists until ~41 T where the magnetization jumps to $M = M_{sat}$. The transitions appear to be of first order, being very sharp and with a clear hysteresis in the pulsed magnetic measurement field. Mean-field theory was used to calculate the magnetic phase diagram and only two possible magnetic structures for the ½ magnetization plateau were determined, neither being compatible with the magnetic exchange parameters determined by neutron spectroscopy [2]. Indeed, the theory showed that the appearance of a ½ magnetization plateau was incompatible with these parameters [4].

The experiment aimed to determine the magnetic structure of the ½ magnetization plateau to test the theory, and to provide further insight into the magnetic Hamiltonian for $FePS_3$. The maximum field of the 40 Tesla pulsed magnet was expected to be sufficient to drive the sample into the plateau state, although probably not into the saturated state.

The previous high-field magnetization experiments used samples cut from a well-characterised single crystal of $FePS_3$ [4]. The experiment used the remainder of the crystal, which was a platelet having an approximately right-angle triangle surface area with dimensions 7×10 mm^2 and a thickness of ~0.2 mm. The normal to the surface was parallel to the **c*** axis, and to the aligned moments in the antiferromagnetic phase.

The sample had to be carefully aligned as the 40 Tesla magnet has relatively narrow entry and exit windows, with ±15° about the field axis on one side and ±30° on the other. The crystal structure for $FePS_3$ has the monoclinic space group $C\frac{2}{m}$ and the magnetic propagation vector in zero field of $\mathbf{k}_M = [01½]$ [2]. The sample had to be mounted with the field normal to the platelet for the **H** || **c*** measurements. The low-symmetry space group, the requirement for the sample orientation with respect to the applied field, the fact that the magnetic propagation vector in zero field has a component along **c*** and the limitations on the beam opening for the magnet severely constrained the regions of reciprocal space with magnetic peaks that could be accessed.

Calculations showed that the 001, $\bar{1}30$ plane and the 001, $\bar{1}40$ plane would be accessible by tilting the magnet about the horizontal field axis by 6.6°, which was acceptable. The 001, $\bar{1}30$ plane has a strong, accessible nuclear peak at $\bar{1}30$ but does not have any magnetic Bragg peaks at H = 0, while the second plane has no nuclear peaks but has an accessible magnetic Bragg peak at $1\bar{4}\frac{\bar{1}}{2}$. Magnetic Bragg peaks appear in both planes for either of the proposed structures ½ magnetization plateaux. Figure 1 shows various examples for the beam and magnet configurations, using an incident beam with $k_i = 4.1$ Å^{-1}. The experiment was performed with a slightly different $k_i = 4.15$ Å^{-1}

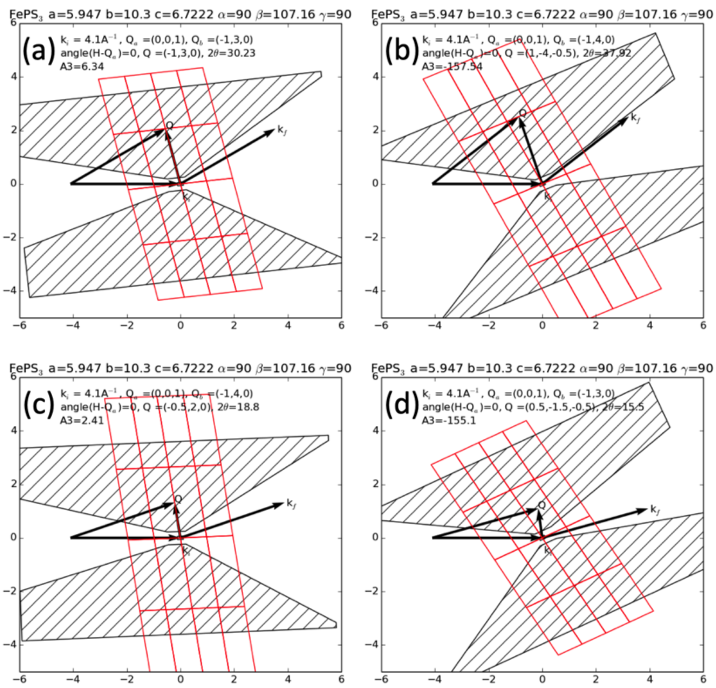

Figure 1: Calculated configurations to access various reciprocal lattice positions for FePS3. (a) the $\bar{1}30$, which a nuclear position and is not magnetic at H = 0 but is potentially magnetic at M/M_{sat} = ½, (b) the $1\bar{4}\frac{\bar{1}}{2}$, which is magnetic-only at H = 0, (c) the $\frac{\bar{1}}{2}20$ and (d) the $\frac{1}{2}\frac{\bar{3}}{2}\frac{\bar{1}}{2}$, which are not present at H = 0 but are potential magnetic peaks at M/M_{sat} = ½. (a) and (d) are in the 001, $\bar{1}30$ plane and (b) and (c) are in the 001, $\bar{1}40$ plane. (a) and (c) have the incident beam entering through the smaller window on the magnet while (b) and (d) have the magnet rotated by 180° to have the incident beam entering through the larger window.

The sample was pre-aligned using IN3 and was glued onto a sapphire support appropriate for the magnet. An immediate problem was identified when the sample was installed on IN22: the mounting used to hold the sapphire support differed between IN3 and IN22. The alignment performed on IN3 was therefore correct with respect to its sample rotation (A3), but the zero differed between IN3 and IN22 and the alignment on IN22 had a global, and unknown, A3 offset.

IN22 was configured without energy analysis to improve statistics. Only one strong nuclear Bragg peak, the $\bar{1}30$, was accessible with the magnet, and considerable time was spent searching for the peak. A candidate peak was found, the crystal was cooled to 1.8 K and a search was conducted for the $1\bar{4}\frac{1}{2}$ but it could not be located. Pulsed-field measurements to 40 Tesla were then started as time was progressing, looking for changes in the $\bar{1}30$ Bragg peak and for a potential peak at $1\bar{3}\frac{\bar{1}}{2}$. No changes were detected. Replacing the energy analysis resulted in the disappearance of the presumed $\bar{1}30$ Bragg peak, showing it to be spurious. Further thought revealed that the wrong handedness had been defined for the crystal axes and that the magnet had to be rotated by 180° for the correct geometry to measure the $\bar{1}30$, as shown in figure 1(a).

A continued search located a peak that was thought to be the $\bar{1}30$. The peak was much weaker than expected, but was at the expected position and was present with the analyser in and at different k_i. This peak was measured with the pulsed field. It did not show an anticipated increase in intensity from a new magnetic order, but did show a decrease as shown in figure 2. The decrease begins at ~10 Tesla and changes steadily and reversibly to the maximum field. Measurements to 20 Tesla at slightly different A3 angles did not show any intensity increase, suggesting that the data in figure 2 show a loss of intensity in the peak and not a shift in its position. Finally, measurements were performed at $1\bar{3}\frac{\bar{1}}{2}$, but no intensity was found at any field.

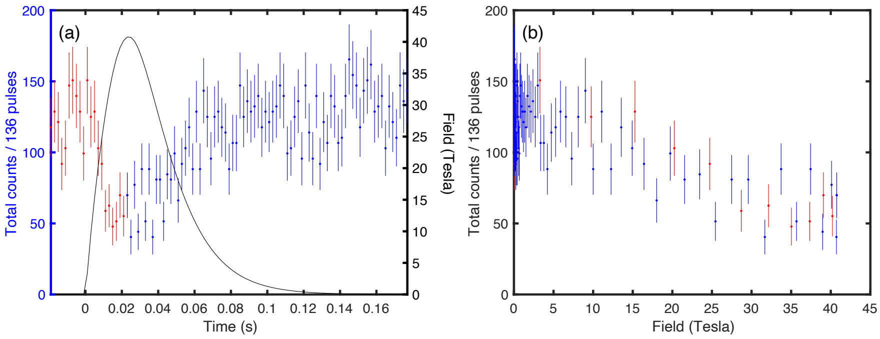

Figure 2: Pulsed field data measured at the $\bar{1}30$ and at 1.8 K. The intensities for rising fields are plotted in red, and for falling fields in blue. (a) Data shown as a function of time, with the field as a function of time plotted with respect to the right-hand y-axis (b) Data shown as a function of instantaneous field.

On opening the magnet, it was found that most of the sample had fallen from the mount and had self-delaminated, coating the inside of the sample space. A small amount of crystal remained attached to its sapphire support, and it appears that the data in figure 2 came from this remanent. $FePS_3$ is known to self-delaminate in large fields due to strong magnetostriction when the field is applied along **b** [4]. While this was nominally orthogonal to the applied field direction, some magnetostriction for fields along **c*** may also occur, and may be a source for the intensity decrease in figure 2. Furthermore, it is likely that there was a misalignment between the field direction and the **c*** axis, not least because of the discrepancy between the A3 zero on IN3 and IN22. This was probably only a few degrees, however the samples used for the magnetometry experiments were substantially smaller. It appears likely that the magnetostriction was sufficiently large on the relatively large sample that the majority detached itself and fell. It is quite likely that this happened early in the experiment, when the pulsed fields were first applied and the sample orientation was incorrect.

In conclusion, the experiment appears to be feasible, but changes must be made if it is to be repeated.

Lessons learned:

- A device to mount the crystal onto the full magnet insert (sapphire support plus sample stick used for the 40 Tesla magnet) must be made and used for checking the sample alignment. The device must have a known orientation with respect to the zero for the A3 axis (or equivalent) on the instrument used to check the alignment. (Ideally, IN22).
- The sample alignment should be checked below the Néel temperature (< 125 K) to identify the $1\bar{4}\frac{\bar{1}}{2}$. In the latter part of the experiment, when the data in figure 2 were measured, it is probable that the Bragg intensity from the crystal remanent was too small to be reliably measured. The peak must be properly characterised outside the magnet, and must be identified and measured inside the magnet before pulsing.
- The positions of the magnet windows with respect to the crystal orientation matter as, due to the monoclinic space group, reciprocal space access is not invariant to a 180° rotation. Great care must be taken that the sample is placed in the correct orientation in the magnet.
- A sapphire sample mount must be made that prevents the sample from falling. Fixing the sample between a sapphire "sandwich" would be an option.

References:

[1] K. Kurosawa *et al*., J. Phys. Soc. Japan **52** (1983) 3919
[2] D. Lançon *et al*., PRB **94** (2016) 214407
[3] K. Okuda *et al*., *High Field Magnetism*, ed. M. Date (North Holland, Amsterdam, 1983) p. 55
[4] A. R. Wildes *et al*., PRB **101** (024415) 2020