Experimental report

05/09/2022

Proposal: **4-01-1608** **Council:** 10/2018

Title: The magnon dispersion in single crystals of NiPS3

Research area: Physics

This proposal is a new proposal

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Samples: NiPS3

Instrument	Requested days	Allocated days	From	To
IN8	8	4	03/07/2019	08/07/2019

Abstract:

NiPS3 is a quasi-2D antiferromagnet with a honeycomb lattice. We have previously attempted to estimate the magnetic exchange interactions from neutron inelastic spectrometry measurements on a powdered sample. The analysis required a number of assumptions and the results were somewhat ambiguous. We now wish to perform more precise measurements on single crystals. Our first aim is to measure the interplanar exchange to determine how close NiPS3 is to a true 2D magnet. We will then measure the in-plane magnon dispersion to verify the magnon dispersion and the magnetic Hamiltonian.

Report for experiment 4-01-1608: The magnon dispersion in single crystals of $NiPS_3$

Experimental team: Andrew Wildes and Ketan Anand

1. *Alignment verification performed on IN3, 14 – 16 June 2019*

Numerous relatively large samples were synthesized. Crystals from this family of materials are known to suffer from significant twinning [1] in the form of three domains rotated by 120° about the **c*** axis. Consequently, careful characterisation of the samples was required to identify the best candidates for the experiment.

Two methods were used to characterise the domain population.

i) The sample was aligned in a nominal 0*kl* plane and cooled to 100 K, below the Néel temperature at ~165 K. The intensity of the magnetic 010 Bragg peak was measured. The sample was then removed, rotated by 60° about the **c*** axis, re-cooled and the 010 peak from the new domain was measured. The process was repeated for the third domain.

ii) The sample was placed on a horizontal mount and then cooled to 100 K. The **c*** axis is vertical in this orientation, and the horizontal scattering plane is 3*h k –l*. The 010 Bragg peaks from the three domains were then all accessible and could be measured without removing the sample from the cryostat.

The best crystal was chosen for the IN8 experiment. The crystal had dimensions of ~$10 \times 10 \times 0.3$ mm^3 and the ratio of domain populations was 0.58:0.26:0.16. The sample was aligned in the 0*kl* scattering plane for the experiment.

2. *Experiment on IN8, 2 – 8 July 2019*

IN8 was configured with the PG002 monochromator and analyser, and 40′ collimators in positions $\alpha_{1..4}$. The instrument resolution was calibrated by measuring the energy width of the incoherent scattering from a single crystal of $MnPS_3$, and by mapping the reciprocal space around its 200 Bragg peak. The sample was cooled to 1.5 K using an orange cryostat.

The 0*kl* scattering plane gave access to Brillouin zone centres at 010 and 011. The spin waves are steeply dispersive in $NiPS_3$ [2] and the experiment focused on the lower energy regime, measuring the scattering up to energy transfers of 21 meV.

Previous measurements on powdered samples [2] suggested that $NiPS_3$ has an energy gap of ~7 meV at the Brillouin zone centre. The IN8 experiment showed weak but clear spectral weight below this energy, revealing a previously undiscovered mode that is steeply dispersive that had no gap within the energy resolution of the measurement. The mode is dispersive along both *k* and *l*. The dispersion along *k* was unsurprising as $NiPS_3$ is a van der Waals compound with the Ni^{2+} ions forming a honeycomb lattice in the *ab* planes. The compound was therefore assumed to be quasi-two-dimensional with very weak magnetic interactions along the **c*** axis. The discovery that the spin waves are strongly dispersive along *l* was therefore surprising, showing that $NiPS_3$ is magnetically more three-dimensional than previously thought.

Figure 1 shows representative constant energy scans parallel to the **b*** axis through the 01*l* positions for different values of *l*. The data at 011 show clear peaks down to 2 meV energy transfer, and the peaks rapidly disappear at $l \neq 1$ for $\Delta E \leq 10$ meV. Complementary measurements were made for trajectories along the **c*** axis, and measurements at 300 K verified that the peaks disappeared above the Néel temperature.

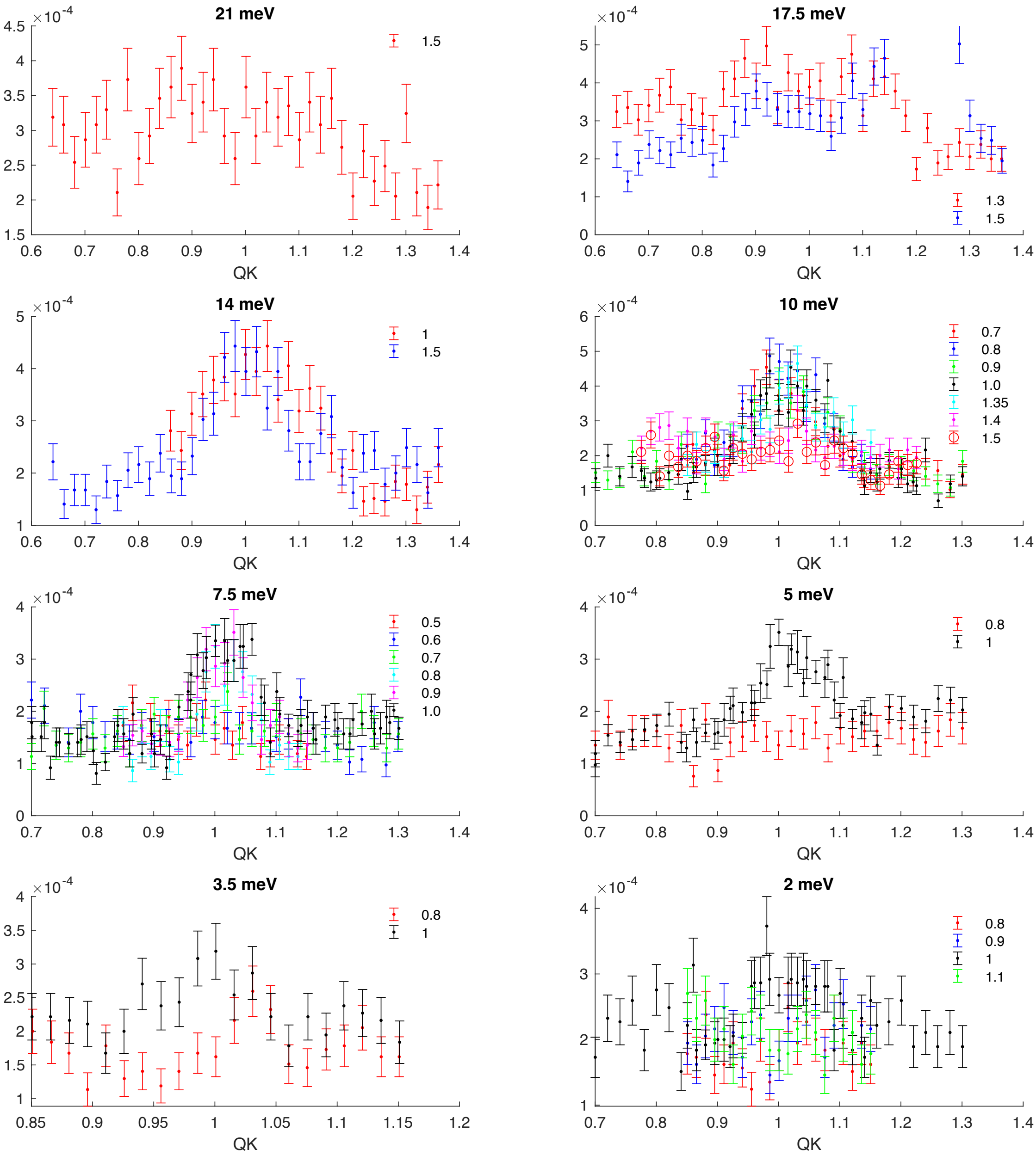

Figure 1: Example measurements of NiPS3 on IN8. The data correspond to constant energy scans parallel to the **b*** axis, centred at 01*l* for various values of *l*.

The data have been subsequently analysed using a Heisenberg Hamiltonian with a combination of a uniaxial and planar single-ion anisotropies, which lifts the degeneracy of the low-energy spin waves at the Brillouin zone centre.

References:

[1] C. Murayama *et al.*, JAP **120** (2016) 142114

[2] D. Lançon *et al.*, PRB **98** (2018) 134414