Experimental report

12/09/2019

Proposal: 4-01-1632 **Council:** 4/2019

Title: The spin wave spectrum of CoPS3

Research area: Physics

This proposal is a new proposal

Main proposer: Andrew WILDES

Experimental team: Bjorn FAK
Luc TESTA
Ursula Bengaard HANSEN

Local contacts: Andrew WILDES

Samples: CoPS3

Instrument	Requested days	Allocated days	From	To
IN8	8	7	11/07/2019	18/07/2019

Abstract:

CoPS3 is a quasi-two-dimensional antiferromagnet with a honeycomb lattice. Recent theory calculations predict that the spin-orbit coupling is large enough to create Kitaev-Heisenberg physics. We wish to use neutron three-axis spectrometry using IN8 to measure the spin wave dispersion in the compound to test the theory, and to compare the spin dynamics of CoPS3 with its sister compounds MnPS3, FePS3 and NiPS3.

$CoPS_3$ is a quasi-two dimensional antiferromagnet [1]. The Co^{2+} ions form a honeycomb lattice in the *ab* planes. The compound orders magnetically below the Néel temperature of 122 K with a zig-zag structure and the moments are approximately collinear with the crystallographic **a** axis. Magnetic susceptibility shows the compound to have a large XY anisotropy with the **c*** axis being the magnetic hard direction.

Our experiment aimed to use neutron three-axis spectrometry to measure the spin waves in a single crystal of $CoPS_3$ to determine the exchange parameters and to elucidate the Hamiltonian. Based on magnetization measurements and experience from modelling the spin wave dispersions in the Mn, Fe and Ni sister PS_3 compounds, we estimated that the spin wave dispersion would be relatively flat with a spectral weight between 20 and 35 meV. Our estimate is shown in Figure 1.

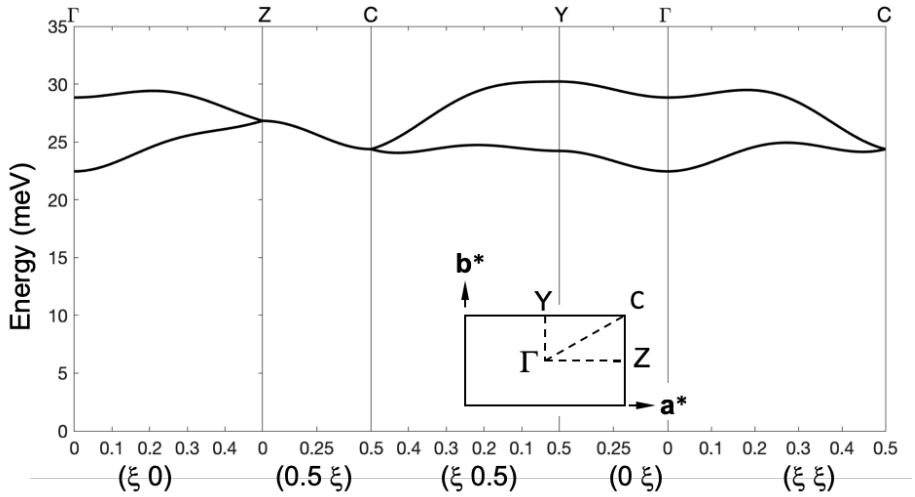

Figure 1: Estimated spin wave dispersion for $CoPS_3$, calculated using a Heisenberg Hamiltonian with a single-ion anisotropy and using values determined from Molecular Field theory for the exchange parameters and anisotropy. The in-plane Brillouin zone is shown in the insert.

We were awarded 8 days of time on IN8. We co-aligned 4 high-quality single crystals for the experiment with a total mass of ~0.05 – 0.1 g. The crystals were glues to an aluminium plate using GE varnish. The transition metal-PS_3 compounds are known to be highly prone to crystal twinning [2], however the crystals chosen for the IN8 experiment were shown to be single domain to better than 75%. The samples were aligned for the experiment with the scattering plane spanned by the **a*** and **b*** axes.

A number of experimental configurations were tested. The two configurations that gave the best data were:

i) Pyrolytic graphite (002) monochromator and analyser, both with vertical and horizontal focusing.
ii) Silicon (111) monochromator and Pyrolytic Graphite (002) analyser, both with vertical and horizontal focusing.

Both configurations were operated with the final wavenumber fixed at $k_f = 2.662$ Å^{-1}. Configuration i) provided better resolution and intensity, however it was prone to spurions. The bulk of the measurement time was spent using configuration ii), which gave a cleaner signal but required longer counting time and had somewhat worse resolution.

The inelastic scattering was measured at numerous points in two Brillouin zones centred at (2 1 0) and (1 4 0) respectively. Measurements were largely performed at 1.5 K, with the

temperature being raised above the Néel temperature to 150 K for measurements during the final day. Excitations were found in the energy transfer region 10 – 30 meV. The excitations were weakly dispersive. Both of these observations were consistent with the calculations.

Figure 2 shows the measured scattering at the (2 1 0) Brillouin zone centre for temperatures of 1.5K and 150 K. Three distinct peaks are observed at 1.5 K with energies of ~15, 19 and 23 meV. Optical phonons are also expected in this energy range [3] and the measurement at 150 K shows that the phonons give a broad peak centred at ~21 meV. The differences between the scattering at the two temperatures suggest that $CoPS_3$ has at least three separate spin wave branches, in contradiction to the predicted spin wave dispersion which consisted of two doubly-degenerate branches. Co^{2+} is known to have a large spin-orbit interaction and it is likely that this lifts the degeneracy of the spin wave energies, hence having more than two observable modes.

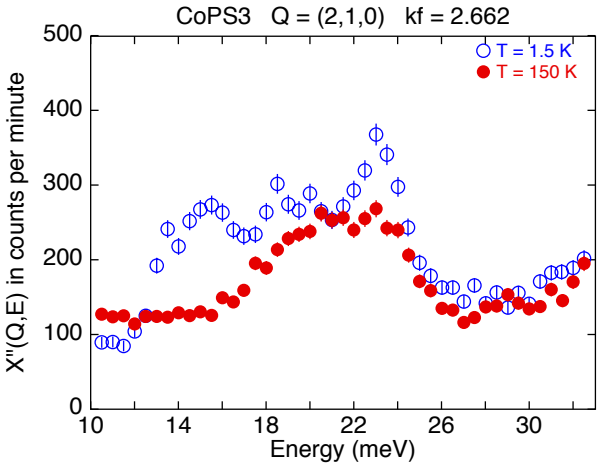

Figure 2: Measurements of the inelastic scattering at the (2 1 0) Brillouin zone centre at 1.5 K (blue points) and 150 K (red points).

Unfortunately, the phonons are at exactly the same energies as the spin waves and they persisted throughout the Brillouin zones. Figure 3 shows data from two points in the Brillouin zone boundary at the two temperatures. Both show that the scattering at both temperatures is dominated by the phonons, however a clear peak at ~27 meV
Is visible in the data at 1.5 K. This peak is most likely due to the spin waves. The low temperature data show hints of other spin wave peaks, however the phonon contribution is too large to be able to state their contribution with any confidence.

The instrumental background was measured by rotating the analyser by 5 degrees and repeating the scans. The background was very low, ~10 counts for equivalent monitor and independent of the energy transfer. The larger apparent background will have a contribution from the incoherent scattering from the GE varnish used to glue the crystals.

A further experiment is required to quantify the spin wave scattering and to be able to model the spin wave dispersion. The experiment must explicitly account for the phonon contribution. Three possibilities are immediately apparent:

i) Further measurements of the scattering at low and high temperatures, and then to perform a temperature subtraction to account for the phonons. This method was

attempted for the data collected in this experiment, although unfortunately insufficient time was allocated to measure data at 150 K and only a limited number of scans could be performed. This method relies on assumptions that the phonon intensity depends purely on the temperature-dependent Bose factor and that the other scattering is directly comparable between the two temperatures. The large difference in the required temperatures (1.5 K to 150 K) may test those assumptions.

ii) To measure the scattering at much larger momentum transfers to estimate the phonon contribution at smaller momentum transfers, and thus to allow the phonons to be subtracted. This method relies on the knowledge of the phonon structure factor, in addition to the expected Q-dependence, and is again prone to systematic errors inherent in the requirement for certain assumptions.

iii) To use neutron polarization analysis to cleanly separate the magnetic and phononic contributions to the scattering. This is by far the best option as it will also account for any nuclear spin-incoherent scattering, although the count times will be long.

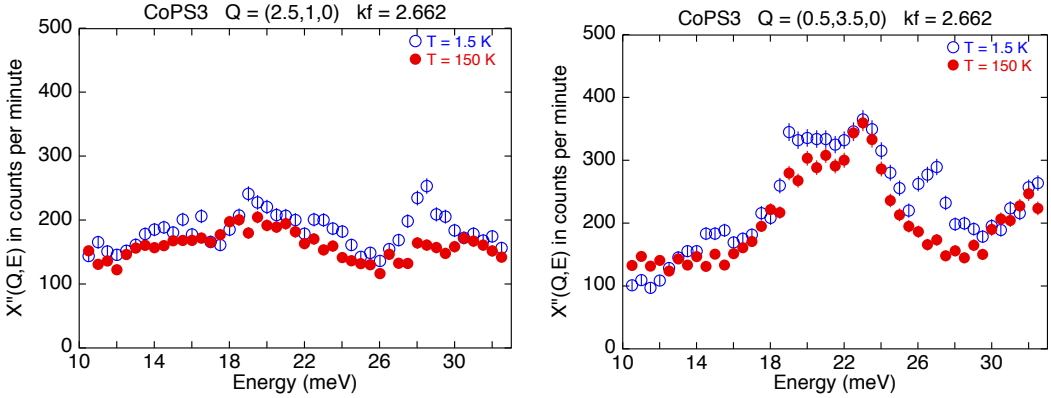

Figure 3: Measurements of the inelastic scattering at (2.5 1 0), corresponding to the Z point in figure 1, and (0.5 3.5 0), corresponding to the C point in Figure 1. Data measured at 1.5 K (blue points) and 150 K (red points) are shown.

References:

[1] A. R. Wildes *et al.*, J. Phys.: Condens. Matter **29** (2017) 455801

[2] C. Murayama *et al.*, J. Appl. Phys. **120** (2016) 142114

[3] M. Bernasconi *et al.*, Phys. Rev. B **38** (1988) 12089