

Experimental report

11/11/2022

Proposal: DIR-248

Council: 4/2021

Title: MAGNETIC EXCITATIONS IN THE QUANTUM SPIN-LIQUID CANDIDATE S=1 KAGOME
ANTIFERROMAGNET ND₄Ni_{2.5}V₂O₇(OD)₂.D₂O

Research area: Physics

This proposal is a continuation of 4-05-827

Main proposer: Eamonn CONNOLLY

Experimental team: Jacques OLLIVIER

Local contacts: Bjorn FAK
Jacques OLLIVIER

Samples: ND₄Ni_{2.5}V₂O₇(OD)₂.D₂O

Instrument	Requested days	Allocated days	From	To
IN5	2	2	12/07/2021	14/07/2021

Abstract:

Experimental report: Magnetic excitations in the quantum spin-liquid candidate $S = 1$ kagome antiferromagnet, $\text{ND}_4\text{Ni}_{2.5}\text{V}_2\text{O}_7(\text{OD})_2\text{D}_2\text{O}$

The aim of this experiment was to probe the magnetic excitations in $\text{ND}_4\text{Ni}_{2.5}\text{V}_2\text{O}_7(\text{OD})_2\text{D}_2\text{O}$, using inelastic neutron scattering (INS) on IN5. $\text{ND}_4\text{Ni}_{2.5}\text{V}_2\text{O}_7(\text{OD})_2\text{D}_2\text{O}$ is a novel $S = 1$ kagome material consisting of isotropic Kagome layers of $S = 1$ Ni^{2+} ions. These are well separated by diamagnetic bivanadate pillars, interstitial ND_4^+ ions and D_2O molecules, in a structural framework similar to the $S = 1/2$ kagome antiferromagnet (KAFM) volborthite. Bulk magnetometry showed an absence of magnetic order down to 5 K, despite an antiferromagnetic field of $\theta_w = -32$ K and curvature in the inverse susceptibility at $T < 150$ K. This absence of magnetic order was further confirmed down to 280 mK through μSR measurements. The disordered-dynamic ground state of $\text{ND}_4\text{Ni}_{2.5}\text{V}_2\text{O}_7(\text{OD})_2\text{D}_2\text{O}$ suggests it may represent a rare example of a quantum spin-liquid (QSL) state in a $S=1$ kagome system.

Our recent inelastic neutron scattering experiment on PANTHER (4-05-807) evidenced low- E low- Q magnetic excitations. An incoming energy of $E_i = 19.03$ meV at $T = 10$ K showed magnetic scattering centered at $Q \sim 1.5 \text{ \AA}^{-1}$. The magnetic origin of the scattering is confirmed by the extracted imaginary magnetic susceptibility against energy at $Q = 1.50 \pm 0.5 \text{ \AA}^{-1}$. The scattering observed at $E < 4$ meV disappears at $T > 50$ K and indicates the onset of magnetic scattering. The observed magnetic scattering formed in the tail of the Bragg peaks making detailed characterisation of the magnetic excitations difficult. For $E_i < 19.03$ meV similar problems were observed, where the reduced flux of PANTHER at low energies was detrimental to more detailed characterisation. In order to complete the characterisation of the low $E - Q$ region directorial discretionary time was awarded for further analysis on the IN5 spectrometer.

For the experiment on IN5, 3.5 g of sample was loaded into a 15 mm diameter aluminium can and cooled to 1.5 K. We collected data with energy $E_i = 3.55$ meV for 2 hours, followed by a further collecting for 2 hours at 50 and 100 K. Magnetic scattering was observed at $T < 50$ K and further scans at 20, 10 and 5 K were completed to track the evolution of the scattering with temperature. The magnetic scattering was isolated by subtraction of the $T = 50$ K data from the $T = 1.5$ K data (Figure 1a). The temperature subtracted data shows a magnetic excitation with a broad Q -range of $Q \sim 0.25 - 2.75 \text{ \AA}^{-1}$. Cuts through the dynamic susceptibility at $Q = 1.2 \text{ \AA}^{-1}$ show the build-up of the excitation at $T < 50$ K with intensity maximum at $E_{\text{max}} = 1.20$ meV (Figure 1b).

The energy was changed to $E_i = 2.27$ meV to explore a potential gapless magnetic excitation at low- Q and low- E . This excitation is not present in temperature subtracted data and is believed to be a temperature-independent spurion. Scans were also completed at $E_i = 10.43$ and 5.11 meV for temperatures between $150 - 1.5$ K to increase the kinematic window and explore the Q dependency of the $E_{\text{max}} = 1.25$ meV magnetic excitation. Further analysis of the Q -dependence and potential gap for this magnetic excitation is ongoing. The combined PANTHER and IN5 data will allow determination of the low-energy profile of the magnetic scattering in $\text{ND}_4\text{Ni}_{2.5}\text{V}_2\text{O}_7(\text{OD})_2\text{D}_2\text{O}$.

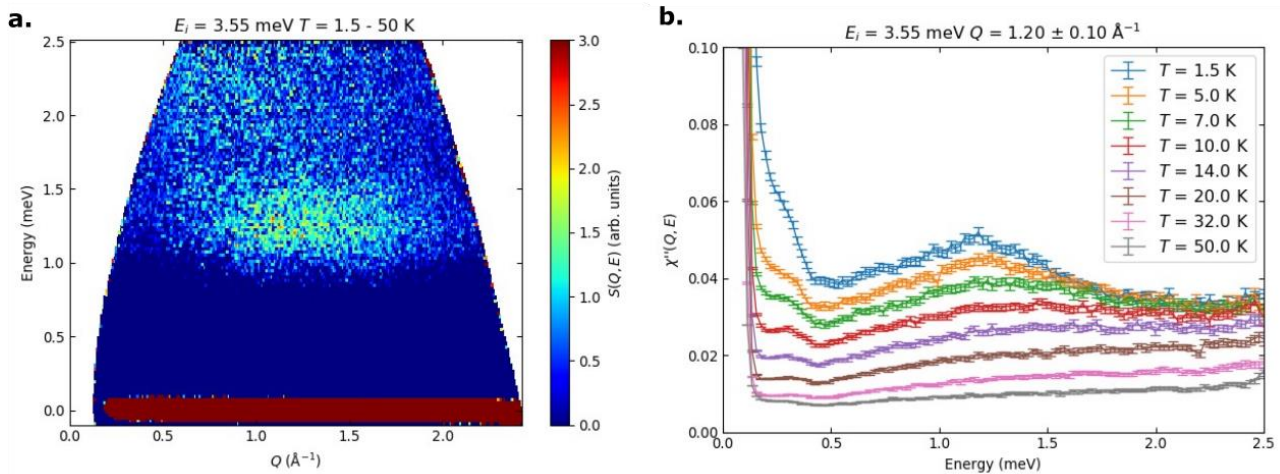


Figure 1 **a.** Temperature subtracted ($T = 1.5 - 50$ K) $S(Q, E)$ map using $E_i = 3.55$ meV data. A gapped magnetic excitation from $Q \sim 0.25 - 2.75 \text{ \AA}^{-1}$ is observed **b.** Temperature dependant Q -cuts of $\chi''(Q, E)$ showing the build-up of the magnetic excitation with a peak in intensity at $E_{\text{max}} = 1.25$ meV.

