Experimental report

10/03/2021

Proposal: **4-05-827**

Council: 10/2020

Title: Magnetic excitations in the quantum spin-liquid candidate S=1 kagome antiferromagnet ND4Ni2.5V2O7(OD)2.D2O

Research area: Physics

This proposal is a new proposal

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Samples: ND4Ni2.5V2O7(OD)2.D2O

Instrument	Requested days	Allocated days	From	To
PANTHER	3	3	19/02/2021	23/02/2021
IN5	3	0		

Abstract:

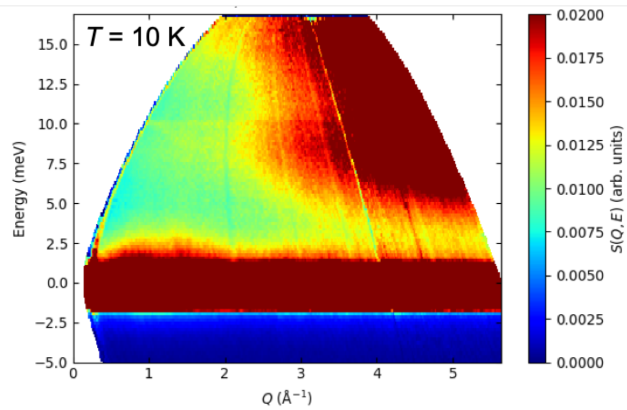
Two-dimensional low-spin kagome magnets are probably the best candidate materials for studies of the novel ground states and exotic, often fractionalized, excitations that characterize quantum spin liquids (QSLs). To date most interest has focussed on S=1/2 QSLs and S=1 systems are poorly understood. We propose to measure the magnetic excitations in the new isotropic S=1 quantum magnet ND4Ni2.5V2O7(OH)2.D2O, which has an ideal (undistorted) kagome lattice and displays spin dynamics and an absence of magnetic order down to 280 mK. The particular aim is to characterise the magnetic excitation spectrum to confirm the ground state character and locate where the system sits in the phase diagram of QSL states.

Experimental report: Magnetic excitations in the quantum spin-liquid candidate $S = 1$ kagome antiferromagnet, $ND_4Ni_{2.5}V_2O_7(OD)_2{\cdot}D_2O$

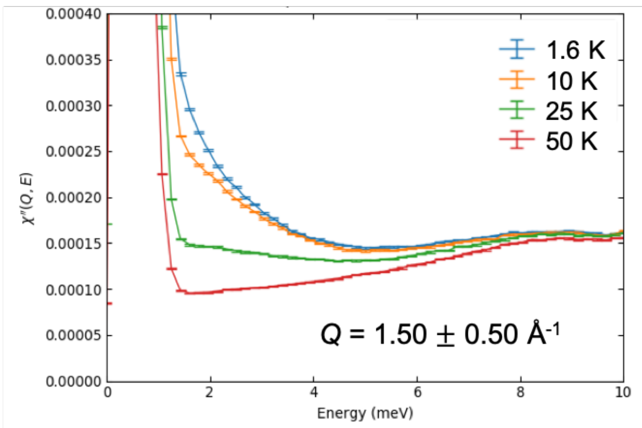
The aim of this experiment was to probe the magnetic excitations in $ND_4Ni_{2.5}V_2O_7(OD)_2{\cdot}D_2O$, using inelastic neutron scattering (INS) on PANTHER. $ND_4Ni_{2.5}V_2O_7(OD)_2{\cdot}D_2O$ is a novel $S = 1$ kagome material consisting of isotropic kagome layers of $S = 1$ Ni^{2+} ions. These are well separated by diamagnetic bivanadate pillars, interstitial ND_4^+ ions and D_2O molecules, in a structural framework similar to the $S = \frac{1}{2}$ KAFM volborthite. Bulk magnetometry showed an absence of magnetic order down to 5 K, despite an antiferromagnetic field of $\theta_W \simeq -32$ 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. In addition, the protonated sample displays a distinct ground state to the deuterated sample, where a transition to a ferromagnetic-like state at $T_C \approx 17$ K is observed in bulk susceptibility. Such distinct ground state properties between the deuterated and protonated samples, induced by small structural changes, suggests this compound lies close to a phase boundary between an ordered state and disordered-dynamic ground state, such as a QSL. Since the energy range of these magnetic excitations was not previously known, a range of incoming energies was required.

A 3.5 g sample was loaded into a 15 mm diameter aluminium can and cooled to 1.6 K. We counted for 3 h with $E_i = 35.00$ meV but it was difficult to find evidence of magnetic scattering, so we measured at base temperature with $E_i = 19.03, 12.50$ and 7.50 meV. We concluded that the magnetic scattering was centred at $Q \sim 1.5\,\text{Å}^{-1}$ and of the order of 1 meV. At 19 meV it was easier to see the magnetic scattering at low-Q compared to the high-Q phonon region. So we did a temperature dependence at 19 meV measuring at 10, 25 and 50 K (4 h runs). Since the magnetic scattering was so low in energy, we did a temperature dependence with $E_i = 12.50$ ($T = 10$ and 50 K) and 7.50 meV ($T = 50$ K). We also intended to measure $E_i = 7.50$ at 10 K but the He dewar ran empty causing the system to rapidly heat up to high temperatures. With $E_i = 12.50$ and 7.50 meV, the flux is reduced by a factor of two so the collection times were between 6 and 7 h per temperature.

At 10 K with $E_i = 19.03$ meV, the dynamic structure factor, $S(Q, E)$, (Fig. 1a) shows evidence of magnetic scattering below ~ 2.5 meV, which becomes more obvious when looking at the temperature dependence in $\chi''(Q, E)$ (Fig. 1b). It is difficult to extract the magnetic scattering as the tail of the elastic peak extends up to ~ 2 meV, so further measurements with lower incoming energies will be required to investigate in detail the magnetic excitations.



(a) $S(Q, E)$ at 10 K with E_i=19.03 meV.



(b) E_i=19.03 meV: $Q = 1.5 \pm 0.5\,\text{Å}^{-1}$ at various T in $\chi''(Q, E)$.