Experimental report

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Title: Excitation spectrum of the kagome-lattice antiferromagnet Nd3BWO9

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Samples: Nd3BWO9

Instrument	Requested days	Allocated days	From	To
IN5	3	6	07/11/2023	13/11/2023

Abstract:

The recently discovered rare-earth based quantum magnet Nd3BWO9 poses a rare realization of the kagome lattice antiferromagnet with effective spin-1/2 degrees of freedom and no structural disorder. It is therefore an ideal candidate in the quest towards quantum spin liquids. Thermodynamic and neutron diffraction experiments suggest a highly frustrated ground state with strong antiferromagnetic correlations but residual magnetic order below 0.3 K due to additional interplane interactions. We hereby propose a single-crystal investigation on the magnetic excitation spectrum of Nd3BWO9, in the search for signatures of fractionalized excitations and proximate spin liquid physics. Due to the suspected continuum excitations at low energy (< 1 meV), the time-of-flight disk chopper spectrometer IN5 will be indispensable for this purpose.

Experimental Report:
Excitation spectrum of the kagome-lattice antiferromagnet Nd_3BWO_9

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The rare-earth based quantum antiferromagnet Nd_3BWO_9 has been proposed [1] as a novel kagome system with effective spin-1/2 degrees of freedom and no structural disorder. It is highly frustrated with $f = T_N/|\theta_{CW}| \gtrsim 10$ and realizes a complex magnetic phase diagram [2]. At zero field, a commensurate-incommensurate transition is found at elevated temperatures, while in applied fields, several magnetization plateau phases with distinct **Q**-modulation are realized. And more recent numerical modeling attempts point to an Ising-like local environment with potentially highly relevant interplane interactions taking the form of frustrated triangular spin-tubes. Given the exotic magnetism and strong frustration, we proposed an investigation of the magnetic excitation spectrum of Nd_3BWO_9 using the time-of-flight disk chopper spectrometer IN5. This should provide valuable insights on the magnetic interactions, constituting a key step in understanding this fascinating quantum material.

340 single crystals were coaligned in the *bc* horizontal scattering plane for a total sample mass of 1.58 g and installed in a dilution refrigerator at $T \sim 40$ mK. Measurements were carried out at $E_i = 1.94$ meV (FWHM resolution $\Delta E = 46$ μeV), rotating 360° in the scattering plane in 2° steps. The powder-averaged neutron spectrum is shown in Fig. 1a, it is dominated by two dispersionless modes at 0.240(2) and 0.306(2) meV, barely visible above the background. This situates Nd_3BWO_9 close to the extreme Ising limit, with fully flat "spin-flip" excitations and a strongly suppressed matrix element (the dominant $|j^z = \pm 9/2\rangle$ ground state weighty cannot be connected by $\Delta S^z = \pm 1$ spin flips). Based on the refined magnetic structure, the two modes can be used to evaluate the dominant exchange parameters as $E_1 = J_1$ and $E_2 = J_1 - J_1' + J_\Delta + J_\nabla$. The dominant couplings turn out to be NN *inter*-plane interactions J_1, J_1' with competing FM-AFM signs – trumping the breathing Kagome exchanges J_Δ, J_∇ (see [3] for details).

Further insights can be gained from the elastic signal (see Fig. 1b): In addition to the known **Q** = (0,0,1/3) bragg peaks, we observe a weak additional modulation **Q** = (1/3,1/3,0). These peaks are at least $\times 20$ weaker than the main order parameter and have not been found in previous diffraction experiments. Moreover, there are strong "planes" of diffuse scattering emanating from the main Bragg peaks perpendicular to l. The associated in-plane and inter-plane correlation lengths $\xi_{ab} \sim 4.6a$ and $\xi_c \gtrsim 78c$ are highly anisotropic, providing further evidence that the magnetism in Nd_3BWO_9 is that of *one-dimensional* Ising spin-tubes, in contrast to previous interpretations in terms of Kagome lattice physics.

Finally, additional data was collected at 0.5 K to characterize the diffuse scattering above T_N. The diffuse planes of magnetic scattering remain present at elevated temperatures, but shifted to incommensurate positions $q_c \approx 0.39$ r.l.u. A complementary analysis to fit the exchange parameters based on molecular dynamics simulations in the paramagnetic state is currently in progress.

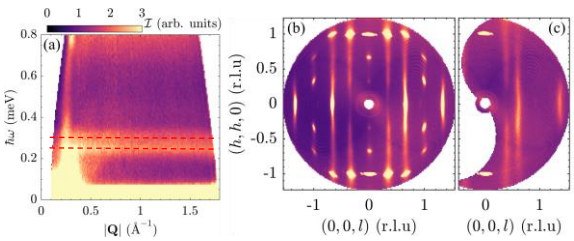

Figure 1: (a) Powder averaged excitation spectrum, composed of two flat modes. (b) The elastic channel exhibits strong diffuse scattering. (c) Addidional data at $T = 0.5$ K, showing persistent incommensurate correlations in the paramagnetic regime.

[1] M. Ashtar et al., Inorg. Chem. 59, 5368 (2020).
[2] D. Flavián et al., PRB 107, 174406 (2023).
[3] J. Nagl et al., arxiv:2409:07807 (2024).