

Experimental report

14/11/2023

Proposal: CRG-3011

Council: 4/2023

Title: Evolution of the magnetic structure of the Kagome-lattice antiferromagnet Nd₃BWO₉ in a magnetic field

Research area: Physics

This proposal is a resubmission of 5-41-1108

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Samples: Nd₃BWO₉

Instrument	Requested days	Allocated days	From	To
D23	5	5	23/08/2023	28/08/2023

Abstract:

The recently discovered kagome-lattice antiferromagnet Nd₃BWO₉ is a promising proximate quantum spin liquid candidate. The crystal field gives rise to anisotropic effective spin-1/2 moments at low temperatures, which exhibit extensive magnetic frustration due to the lattice topology. Bulk characterization reveals residual magnetic order below 300 mK and a complex phase diagram in field, including a fractional $m = 1/3$ magnetization plateau. The propagation vectors have been determined in preliminary neutron diffraction experiments. We hereby propose to refine the magnetic structures via single crystal neutron diffraction, both at zero field and in the partially magnetized plateau phase.

Experimental Report: Evolution of the magnetic structure of the Kagome-lattice antiferromagnet Nd_3BWO_9 in a magnetic field

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Nd_3BWO_9 is a rare-earth based quantum antiferromagnet realizing a breathing kagome geometry in the crystallographic ab -plane [1]. Although additional interplane interactions are likely of relevance, it is highly frustrated, ordering only below $T_N = 0.3$ K. A complex magnetic phase diagram is realized with highly non-trivial spin structures [2]. At zero field an incommensurate state is stabilized, locking in at $\mathbf{Q} = (0,0,1/3)$ at low temperatures. And in applied fields, fractional $m=1/3$ magnetization plateaux are found, with completely distinct modulations $\mathbf{Q} = (0,1/2,1/2)$ and $\mathbf{Q} = (1/3,1/3,1/3)$ depending on the field direction. While these propagation vectors already provide valuable insights, it was not possible to uniquely identify the magnetic structures based on preliminary data. Therefore, we proposed a refinement on single crystals using the thermal lifting counter diffractometer D23, which should provide valuable insights on the magnetic interactions in Nd_3BWO_9 , particularly the role of interplane correlations play in releasing frustration and stabilizing long-range order.

A 17 mg single-crystal was mounted on a copper holder and cooled in a dilution refrigerator to $T = 50$ mK. The sample was aligned with the c -axis vertical and a 6 T cryomagnet was employed to access the in-field phases. A wavelength of $\lambda = 1.28$ Å was chosen and data were collected at various field and temperature conditions. Some preliminary refinement data is shown in Figure 1.

The nuclear structure was determined at 10 K to high accuracy using ~ 500 reflections. We discovered that Nd_3BWO_9 is prone to merohedral twinning: The data could only be refined in the correct space group by including a perfect merohedral twin with permuted reflections $(k,h,-l)$, resulting in an excellent fit $R_F = 2.2\%$.

Next up, ~ 250 reflections were collected at base temperature at the fractional $\mathbf{Q} = (0,0,1/3)$ positions to investigate the zero field magnetic structure. A semi-classical model consistent with symmetry analysis has been

proposed, resulting in a spin density wave structure. It gives decent agreement $R_F = 19\%$ with experiment, though the precise moment modulation is still under investigation.

Finally, a magnetic field $\mu_0 H = 0.85$ T was applied and ~ 180 reflections at $\mathbf{Q} = (1/3,1/3,1/3)$ and $\mathbf{Q} = (1/3,1/3,-1/3)$ were collected. This structure is highly complex, with 9 free refinement parameters even after symmetry constraints. These data are currently being analyzed. In addition to attempts at Rietveld refinement for all distinct structure types, Monte Carlo simulations are being employed to reproduce the observed ordering patterns. An in-depth understanding of the realized magnetic structures will be of key importance in elucidating the magnetism in Nd_3BWO_9 .

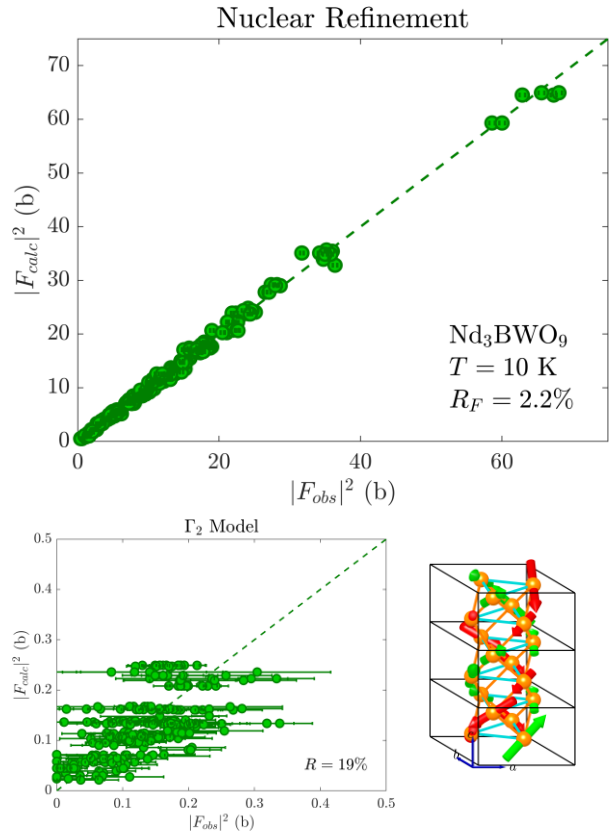


Figure 1: (a) Rietveld refinement of nuclear structure, including a merohedral twin. (b) Preliminary refinement of zero field magnetic structure and corresponding spin configuration.

- [1] M. Ashtar et al., Inorg. Chem. 59, 5368 (2020).
[2] D. Flavián et al., PRB 107, 174406 (2023).