Experimental report

23/08/2022

Proposal:	**DIR-244**		**Council:**	4/2021
Title:	Investigation of magnetic phase transitions in the triangular latticeantiferromagnet Ca3NiNb2O9			
Research area:	Materials			

This proposal is a new proposal

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Samples: Ca3NiNb2O9

Instrument	Requested days	Allocated days	From	To
D10	8	4	20/09/2021	24/09/2021

Abstract:

Triangular lattice Antiferromagnets (TLAFs) exhibit intricate interplay of geometrical frustration, low dimensionality and small spin size leading to exotic magnetic ground states. Ca3NiNb2O9 has a triangular lattice with an isosceles motif, meaning that more exchange interactions are at play. This brings about at least 2 different magnetic order parameters, close-by in temperature. Further, we have succeeded in growing two distinctly different types of single crystalline Ca3NiNb2O9: as-grown and air-annealed and we propose here to study their magnetic ground state. Upon a successful experiment we plan to study an even more interesting high magnetic field phase (at H=10 T) in the near future.

Experimental Report
Investigation of magnetic phase transitions in the triangular lattice antiferromagnet $Ca_3NiNb_2O_9$

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Abstract

Triangular lattice Antiferromagnets exhibit intricate interplay of geometrical frustration, low dimensionality and small spin size, leading to exotic magnetic ground states. $Ca_3NiNb_2O_9$ has a triangular lattice with an isosceles motif, which means more exchange interactions are at play. This brings about at least 2 different magnetic order parameters, close-by in temperature. Two distinctly different types of single crystalline $Ca_3NiNb_2O_9$ were grown: as-grown and air-annealed, and the magnetic structure of the as-grown translucent sample could be studied at D10 and CYCLOPS. Upon this experiment, the even more interesting high magnetic field phase (at $H = 10\,\mathrm{T}$) would provide more insight of the compound.

Single crystals of the compound $Ca_3NiNb_2O_9$ are grown using the four-mirror optical float-zone furnace. For the as-grown translucent (AGT) sample, various reciprocal lattice *hkl*-scans were carried out at D10 at $T = 2\,\mathrm{K}$. It was further examined by measuring Laue patterns at CYCLOPS at $T = 1.5\,\mathrm{K}$ and $10\,\mathrm{K}$ as shown in Fig. 1. Compared to the paramagnetic phase at $10\,\mathrm{K}$, new reflections appear at the base temperature $T = 1.5\,\mathrm{K}$ where the system is magnetically ordered. The patterns could be fitted with the monoclinic space group $P12_1/c1$ within the ESMERALDA software. The magnetic fitting in ESMERALDA supports $\vec{k} \approx (0.046, 0.34, 0)$. However, the broad spots due to the crystal quality (multigrains, as discussed in a previous section) suggest that a simpler commensurate propagation vector of $\vec{k} = (0, 1/3, 0)$ is also plausible.

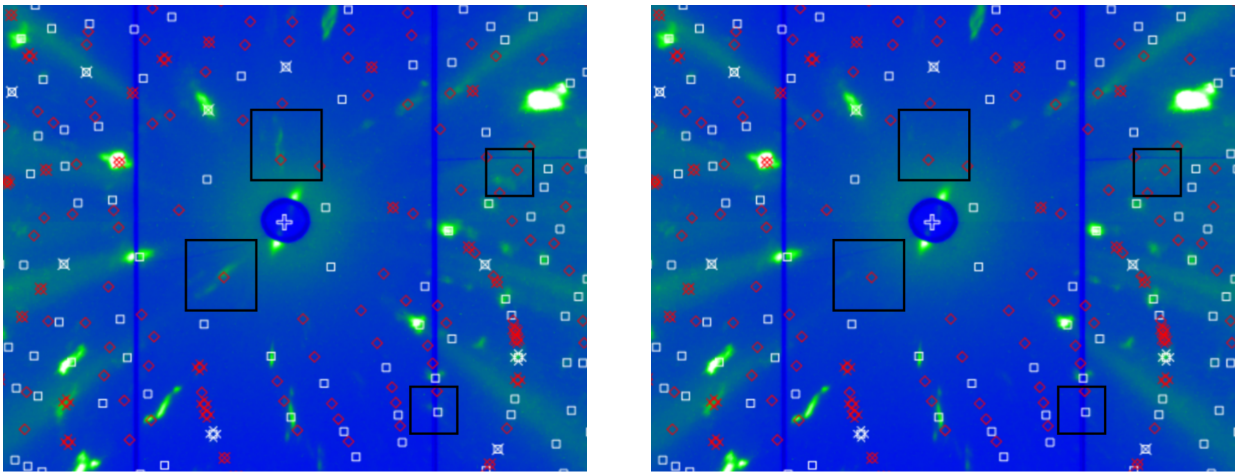

Figure 1: Laue diffraction patterns at $\omega = 0°$, zoomed-in at the centre, at $1.5\,\mathrm{K}$ (left) and $10\,\mathrm{K}$ (right). The white boxes show the fitted nuclear fitting of the monoclinic space group $P12_1/c1$ and the red ones show the magnetic fitting with $\vec{k} = (0.046, 0.34, 0)$. The black frames mark some of the new reflections at $1.5\,\mathrm{K}$ compared to $10\,\mathrm{K}$.

Further examination was performed by measuring different magnetic reflections based on $\vec{k} = (0.046, 0.34, 0)$ at the base temperature $T = 2\,\mathrm{K}$ at D10. The ω-angle denoting sample rotation was scanned around the reflections, with two such representative scans shown in Fig. 2. As visible in the figure, these scans show slightly different peak positions on the position-sensitive detector (± 0.05 for *hkl* in r.l.u.), for the above-mentioned reasons. Although the sample quality did not allow the full determination of the magnetic structure, one could observe strong magnetic satellites around the $(1, 0, 0)$ fundamental reflection and zero intensity of the satellites around the $(2, 0, 0)$, $(0, 2, 0)$ and $(0, 0, 2)$ reflections. This indicates the magnetic arrangement where the moments of Ni atoms located on the 2a-site, $(0, 0, 0)$ and $(0, 1/2, 1/2)$, are ferromagnetic but are antiferromagnetic to those located on the 2d-site, $(1/2, 1/2, 0)$ and $(1/2, 0, 1/2)$, with the moments in the bc-plane.

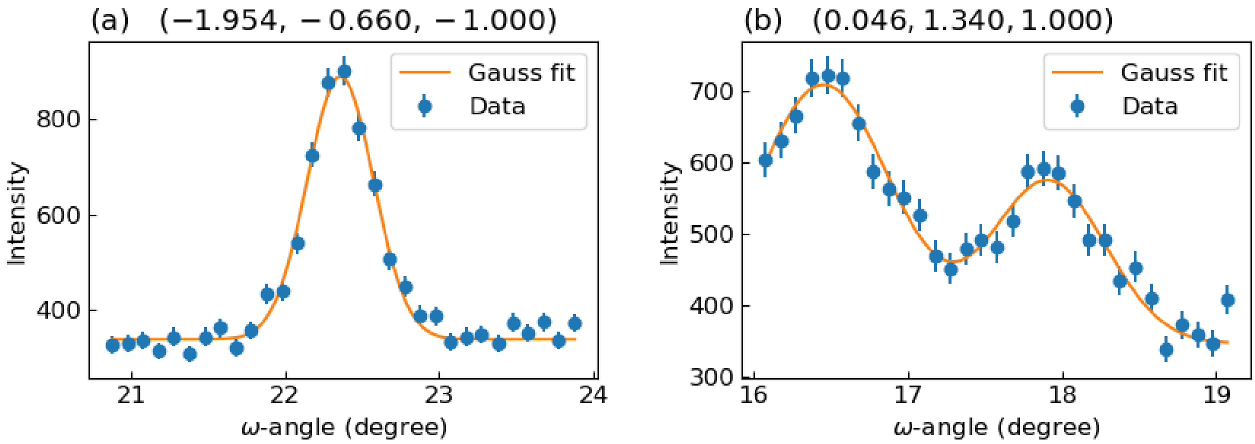

Figure 2: ω-scans around magnetic reflections. Fig.(a) (no twining visible) is fitted with one Gaussian function, whilst Fig.(b) with twinning is fitted with two functions.

The temperature-dependence of the magnetic reflections based on $\vec{k} = (0.046, 0.34, 0)$ was measured at D10, which could be fitted to the power law [1] with a two-step transition

$$I = \begin{cases} a_2 \left(T_{N_2} - T\right)^{2\beta_2} + a_1 \left(T_{N_1} - T_{N_2}\right)^{2\beta_1} + I_b, \ T < T_{N_2} \\ a_1 \left(T_{N_1} - T\right)^{2\beta_1} + I_b, \ T_{N_2} \leq T < T_{N_1} \\ I_b, \ T \geq T_{N_1} \end{cases}, \tag{1}$$

where a_1 and a_2 are scale prefactors, β_1 and β_2 the critical exponents, $T_{N_1} > T_{N_2}$ the transition temperatures, and I_b the background intensity. The least-square fit with this two-step transition gives a better result (χ^2) than with a single-step transition. Two representative scans are shown in Fig. 3, illustrating $T_{N_1} = 4.6(1)\,\mathrm{K}$ and $T_{N_2} = 4.0(1)\,\mathrm{K}$. This agrees with the transition temperatures depicted by the measurement of magnetic susceptibility.

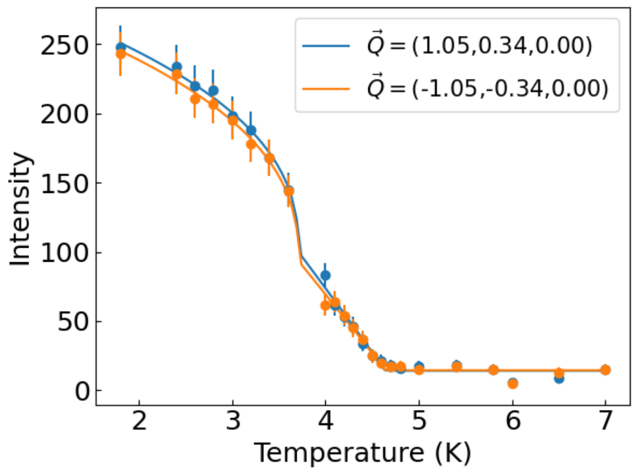

Figure 3: Temperature-dependence scans fitted with a power law as described in Eq. 1. The dots with the error bar show the measured data and the lines of the same colour denote the corresponding fit.

References

[1] Collins, M. *Magnetic critical scattering* (Oxford University Press, New York, 1989).