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Ordered and disordered variants of the triangular lattice antiferromagnet Ca3NiNb2O9

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Single crystals of the triangular lattice antiferromagnet (TLAF) $Ca_3NiNb_2O_9$ and its non-magnetic analogue $Ca_3MgNb_2O_9$ are grown using the four-mirror optical float-zone furnace. During the growth of $Ca_3NiNb_2O_9$, the crystal boule tends to develop cracks upon cooling due to a high-temperature structural modification. Thus, depending on the growth conditions, the crystal boules contain varying amounts of high and low-temperature modifications, present in the form of mm-size grains distinguishable on the basis of their appearance: opaque, dark-green (AGO) and translucent, light-green (AGT) for the high and low-temperature modifications, respectively. Furthermore, when the as-grown, AGO specimen is subject to air annealing at 1200℃, its appearance change from opaque to translucent green, without any noticeable change of weight. Low temperature specific heat and low-field magnetization measurements carried out on the AGO and AGT samples revealed contrasting ground state properties. While AGO exhibits a spin-glass-like ground state, the AGT sample exhibit a two-step, long-range antiferromagnetic ordering of the Ni spins with transitions at $T_{\mathrm{N}_1} =$ 4.6 K and $T_{\mathrm{N}_2} =$ 4.2 K. Detailed structural analysis shows that AGO and AGT crystals crystallize in *Pbnm* (orthorhombic) and $P12_1/c_1$ (monoclinic) space groups, respectively. The high-resolution TEM images confirms the 1:2 ordering of Ni and Nb in the AGT sample. The high-field magnetization up to 50 T in AGT reveals the presence of magnetization plateaus characteristic of TLAFs. The propagation vector in the ordered phase (2 K) is inferred to be $\vec{k} \approx (0, 1/3, 0)$ based on the magnetic neutron scattering.

Primary authors: Mr OULADDIAF, Bachir (Institut Laue-Langevin); Ms ROUT, Dibyata (Indian Institute of Science Education and Research); SKOULATOS, Markos (TUM); TANG, Ran (Technical Univertsity Munich); Mr SINGH, Surjeet (Indian Institute of Science Education and Research)

Co-authors: MIYAKE, Atsushi (The University of Tokyo); TOKUNAGA, Masashi (The University of Tokyo); MAHAPATRA, Sagar (Indian Institute of Science Education and Research); KINOSHITA, Yuto (The University of Tokyo)

Presenter: TANG, Ran (Technical Univertsity Munich)

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