Experimental report

12/10/2020

Proposal: **CRG-2725** **Council:** 10/2019

Title: Crystal field level transitions infrustrated Tb2NbScO7 pyrochlore prone to charge disorder

Research area:

This proposal is a new proposal

Main proposer: **Virginie SIMONET**

Experimental team:
Virginie SIMONET
Elsa LHOTEL
Yann ALEXANIAN
Rafik BALLOU

Local contacts:
Jean Marc ZANOTTI
Quentin BERROD

Samples: Tb2NbScO7

Instrument	Requested days	Allocated days	From	To
IN6-SHARP	5	5	24/08/2020	27/08/2020

Abstract:

EXPERIMENT N° 5-31-2717, CRG-2725 INSTRUMENT: D1B & IN6/SHARP

DATES OF EXPERIMENT D1B : 19/08/2020 → 21/08/2020, IN4C : 24/08/2020 → 27/08/2020

TITLE:

D1B: Charge and magnetic frustration investigated by neutron diffraction in Tb2NbScO7 pyrochlore. IN6/SHARP/ Crystal field level transitions infrustrated Tb2NbScO7 pyrochlore prone to charge disorder

EXPERIMENTAL TEAM: (names and affiliation)

Yann ALEXANIAN, Rafik BALLOU, Elsa LHOTEL, Virginie SIMONET, Sophie deBRION, Institut Néel, CNRS, Grenoble

LOCAL CONTACTS: Claire COLIN

Date of report: 12 October 2020

Pyrochlore compounds are the archetype of 3-dimensional frustrated materials with a plethora of exotic behaviors (spin ice, fragmentation, quantum spin liquid, order-by-disorder) depending on the rare-earth and on the transition metal residing on the two interpenetrated pyrochlore (corner-sharing tetrahedra) sublattices. One of the most intriguing members of the family is $Tb_2Ti_2O_7$ with magnetic rare-earth Tb^{3+}. Instead of displaying the expected Ising antiferromagnetic order, it presents a puzzling correlated state down to the lowest temperature [1]. Additional effects, such as quadrupolar degrees of freedom, must therefore be at play, and enhance quantum fluctuations.

A side route to investigate the origin of the ground state of this material is to change the metal (ex. Sn and Ir in place of Ti) in order to destabilize the fluctuating state and to reveal hidden ingredients of the Hamiltonian [2,3]. A new proposal is to study Niobate compounds where the Nb can be displaced from their ideal position leading to charge correlations, and can be mixed with another transition metal ion [4]. In Nd_2NbScO_7 for instance, the Nb/Sc disorder seems to lead to frustrated electric dipoles mimicking the spin ice state [5]. This disorder nevertheless does not affect much the ground state in this case. As previously observed with Ti, Sn, Hf or Ge substitutions, the Tb based compounds are much more sensitive to the nature of the metal, and the presence of disorder on these sites should considerably modify the stabilized ground state.

We have thus started an extensive study of the sister compound Tb_2NbScO_7 properties, in order to evaluate the role of disorder on its ground state. Moreover, the coexistence of magnetic moments on one pyrochlore lattice and of dielectric moments on the other one might lead to interesting magnetoelectric effects in these materials.

Using a solid-state synthesis method, we have synthesized a polycrystalline sample of Tb_2NbScO_7 with a site mixing of Nb^{5+} and Sc^{3+} on one pyrochlore sublattice.

Experiment N° 4-01-1380

The sample was characterized by X-ray diffraction and magnetometry. The magnetization measurements versus temperature do not reveal any anomaly down to about 1 K where a separation between the field-cooled and zero-field-cooled measurements is observed due to the magnetic interactions between the Tb. We performed a neutron diffraction experiment using a dilution insert on D1B in order to check the ground state of our sample (see figure 1). We observed the absence of magnetic Bragg peaks indicating no long-range order. Instead, a diffuse signal was observed in the difference between 50 mK and 60 K, which is very similar to the one measured in $Tb_2Ti_2O_7$ [1], and was interpreted as a spin-liquid signature.

We performed another experiment using a dilution insert on SHARP to measure the transitions between the crystal field (CF) levels of Tb^{3+} by inelastic neutron scattering. Surprisingly, we observe a shoulder at low energy and low temperature that transforms into a quasielastic signal while heating, but no other excitations that could correspond to crystal field level transitions (see Figure 2). We also checked this absence of inelastic signal on the negative energy transfer side at 200 K. This is at variance with the other Tb pyrochlores where a first crystal field level, responsible for the Ising character of the Tb^{3+}, is observed around 1 meV, and others are visible at higher energies. This absence of inelastic signal could be due to the Nb/Sc disorder, which would broaden the CF levels as in Nd_2NbScO_7. Note however that they are broadened but still visible in this sister compound [5]. It could alternatively be a consequence of the charge distribution (disordered Nb^{5+}/Sc^{3+}) around the Tb^{3+} ions shifting the CF transitions to higher energies.

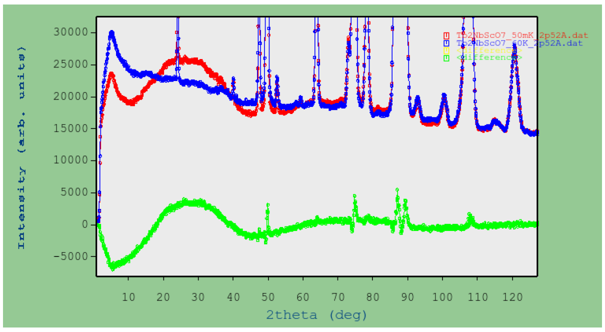

Fig. 1: Powder diffractograms of Tb_2NbScO_7 measured on D1B at 60 K (blue) and 50 mK (red) and there difference (in green) showing the spin liquid like magnetic diffuse scattering.

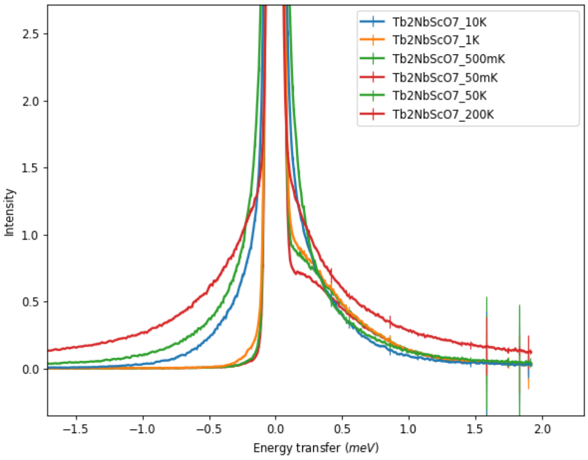

Fig. 2: Thermal variation of the energy spectra of Tb_2NbScO_7 measured on IN6/SHARP at Q=1.2 Å^{-1}.

References:

[1] J. S. Gardner *et al*. Phys. Rev. B 64, 224416 (2001).

[2] I. Mirebeau *et al*. Phys. Rev. Lett. 94, 246402 (2005).

[3] E. Lefrançois *et al*. Phys. Rev. Lett. 114, 247202 2015).

[5] T. M. McQueen *et al.* J. Phys.: Condens. Matter **20**, 235210 (2008).

[6] C. Mauws *et al*., arXiv:1906.10763.