Experimental report

14/03/2021

Proposal: **5-32-911** **Council:** 4/2020

Title: Magnetic diffuse neutron scattering study on quantum spin liquid candidate Ba4NbIr3O12.

Research area: Physics

This proposal is a new proposal

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Samples: Ba4NbIr3O12

Instrument	Requested days	Allocated days	From	To
D7	5	5	03/02/2021	08/02/2021

Abstract:

A new Iridate material Ba4NbIr3O12 has been reported to be a quantum spin liquid (QSL) candidate very recently based on macroscopic thermodynamic measurements. In this frustrated oxide, no long-range magnetic order is observed down to 360 mK. In addition, a low temperature linear behaviour in the specific-heat data strongly advocates for a possible quantum spin-liquid ground state. A frustration parameter of larger than 37 strengthens the possibility of having a quantum spin liquid state furthermore. To observe microscopic evidence of the QSL state, we propose to perform magnetic diffuse scattering experiment with polarized neutron and XYZ polarization analysis on the powder sample. To ensure the quantum behaviour, we propose to perform this measurement in the mK regime using a dilution refrigerator cryostat. This experiment will provide us information about the temperature dependence of the short-range magnetic order, a very important signature in the context of QSL physics. We will also do model calculations with the experimental data to have quantitative idea of the spin Hamiltonian.

<u>Experimental Report</u>

Proposal number: 5-32-911

Title: Magnetic diffuse neutron scattering study on quantum spin liquid candidate $Ba_4NbIr_3O_{12}$.

Aim of the proposed experiment: A new Iridate material $Ba_4NbIr_3O_{12}$ has been reported to be a quantum spin liquid (QSL) candidate very recently based on macroscopic thermodynamic measurements [1-3]. $Ba_4NbIr_3O_{12}$ is a material with a triangular planar geometry of Ir_3O_{12} trimers. This insulating compound crystallizes in a rhombohedral structure (space group *R*-3*m*). Specific-heat measurements performed down to 350 mK at zero magnetic field clearly show absence of any long-range order down to the lowest measured temperature. Furthermore, one can observe a linear temperature dependence in the low-temperature regime of the specific-heat data. Such a behaviour is a promising indication of a possible spin liquid character [4]. A frustration parameter of larger than 37 strengthens the possibility of having a quantum spin liquid state furthermore. To observe microscopic evidence of the QSL state, we proposed to perform magnetic diffuse scattering experiment with polarized neutron and XYZ polarization analysis on the powder sample. To ensure the quantum behaviour, we proposed to perform this measurement in the mK regime using a dilution refrigerator cryostat. With the help of this experiment, we attempted to gather information of the temperature dependence of a possible short-range magnetic order, a very important signature in the context of QSL physics. This would also help to perform model calculations with the experimental data in order to get quantitative idea of the spin Hamiltonian.

Fig-1: *The copper sample holder with annular geometry used to mount the powder sample in our D7 experiment.*

Results obtained from the experiment: To perform this measurement, 5 days on the D7 instrument were allotted. The first day was dedicated to sample installation and instrument calibrations (quartz and vanadium). The remaining days were used for collecting data at different temperatures. We used XYZ polarization analysis available on D7 to separate the magnetic signal from the nuclear one and to determine it in absolute units. As the Ir is a neutron absorber, we used an annular geometry for the Cu sample holder (Fig. 1) to ensure the usage of optimum amount of sample in maximizing the signal. Powder sample of about 2 g mass was used for the measurement. We chose to work with the 4.8 Å wavelength of the incident neutron beam.

Using the dilution fridge insert, the sample was cooled down to 50 mK and the data were collected subsequently. To ensure whether we see a magnetic contribution in the diffuse scattering signal at the base temperature, we heated the system up to 50 K to collect the data at the paramagnetic phase. However, we did not observe any evolution of the magnetic signal between 50 K and 50 mK when compared both the data. To be more conclusive, we further warmed the sample up to 200 K and collected the data. As can be seen from Fig. 2, the magnetic diffuse diffractograms recorded at 50 mK, 50 K, and 200 K look practically the same meaning that there is no magnetic signal present. This is possibly due to the absence of any magnetic order down to 50 mK. However, as the expected moment for Ir is also very weak, it is also possible that the magnetic contribution was too weak to be detected from a powder sample.

In order to overcome this possibility and to reach a conclusion, we plan to use several co-aligned single crystals as the the next step to maximise the signal. It will help observing any magnetic contribution in specific regions of the reciprocal space. We submitted a proposal to D7 for the upcoming cycle to execute this plan with co-aligned single crystals.

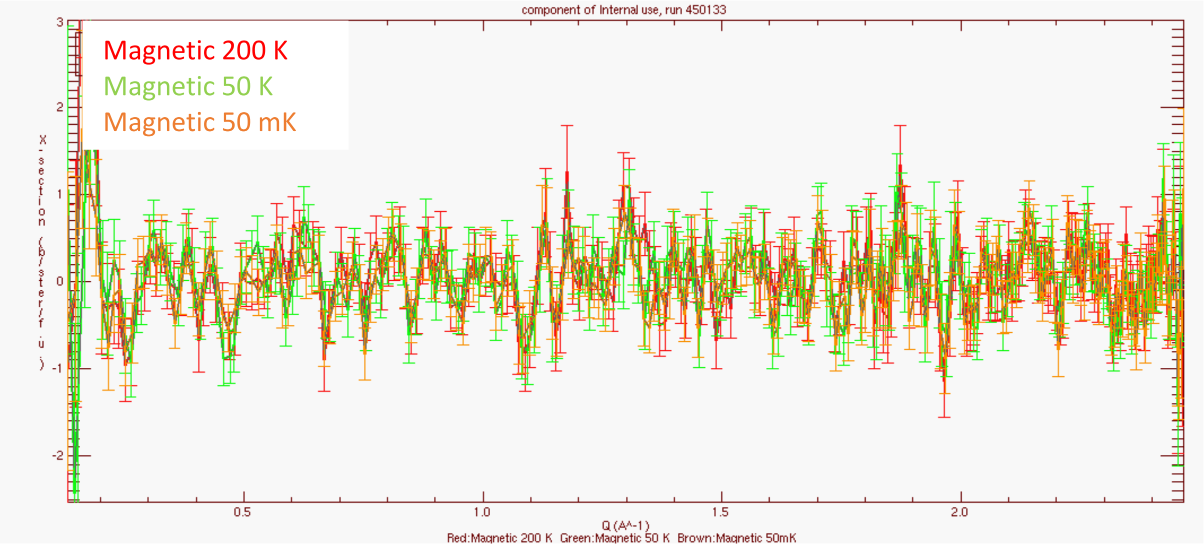

Fig-2: *Magnetic diffuse scattering signal measured on $Ba_4NbIr_3O_{12}$ powder at the D7.*

References:

[1] L. T. Nguyen et al., Phys. Rev. Mater. 3, 014412 (2019);
[2] C. Broholm et al., Science 367, eaay0668 (2020);
[3] G. S. Thakur et al., Cryst. Growth Des. 20, 2871 (2020);
[4] S. Yamashita et al., Nature Phys. 4, 459 (2008).