

# Experimental report

12/09/2023

**Proposal:** 4-01-1759

**Council:** 10/2022

**Title:** Investigation of antiferromagnetic magnon in VBr<sub>3</sub>

**Research area:** Physics

**This proposal is a new proposal**

**Main proposer:** Milan KLICPERA

**Experimental team:** Milan KLICPERA

**Local contacts:** Martin BOEHM

**Samples:** VBr<sub>3</sub>

Instrument	Requested days	Allocated days	From	To
THALES	7	6	24/05/2023	30/05/2023

## Abstract:

Magnetic van der Waals (vdW) crystals show promising potential for high-tech magnetic, magneto-electric, and magneto-optic applications. The discovery of 2D ferromagnetism in a monolayer CrI<sub>3</sub> [1] at elevated temperatures has greatly promoted research in these materials. The large family of transition metal halides offers a large playground for systematic theoretical and experimental investigation of 2D magnetism.

At present, we focus on the antiferromagnet VBr<sub>3</sub>, a member of the trihalide family in which intrinsic ferromagnetism in the single layer limit was discovered in CrI<sub>3</sub>, VI<sub>3</sub>, and even CrBr<sub>3</sub>. VBr<sub>3</sub> magnetic structure also consists of ferromagnetic layers, which are, however, antiferromagnetically coupled through a weak vdW bond. Our very recent Raman spectroscopy experiment has elated antiferromagnetic magnon in VBr<sub>3</sub> at the energy similar (smaller) to that of FM counterparts. The proposed experiment aims just to this magnetic excitation. That is, the dispersion of this magnetic excitation in reciprocal space is to be followed to prove its nature. The results will be subsequently discussed in the frame of other vdW materials.

## Experimental report

<u>Experimental title:</u>	<b>Investigation of antiferromagnetic magnon in VBr<sub>3</sub></b>
<u>Proposal number:</u>	<b>4-01-1759</b>
<u>Instrument:</u>	<b>ThALES</b>
<u>Date of experiment:</u>	24. – 30.5. 2023
<u>Local contact:</u>	Martin Boehm
<u>Experimental team:</u>	Milan Klicpera
<u>Affiliation:</u>	Charles University in Prague, Department of Condensed Matter Physics, Ke Karlovu 5, 121 16 Prague 2, Czech Republic.

### Abstract:

Magnetic van der Waals (vdW) crystals show promising potential for high-tech magnetic, magnetoelectric, and magneto-optic applications. The discovery of 2D ferromagnetism in a monolayer CrI<sub>3</sub> [B. Huang, et al., Nature 546, 270 (2017)] at elevated temperatures has greatly promoted research in these materials. The large family of transition metal halides offers a large playground for systematic theoretical and experimental investigation of 2D magnetism.

At present, we focus on the antiferromagnet VBr<sub>3</sub>, a member of the trihalide family in which intrinsic ferromagnetism in the single-layer limit was discovered in CrI<sub>3</sub>, VI<sub>3</sub>, and even CrBr<sub>3</sub>. The VBr<sub>3</sub> magnetic structure also consists of ferromagnetic layers which are, however, antiferromagnetically coupled through a weak vdW bond. Our very recent Raman spectroscopy experiment has elated the antiferromagnetic magnon in VBr<sub>3</sub> at an energy similar (smaller) to that of its FM counterparts. The proposed experiment aims just at this magnetic excitation. That is, the dispersion of this magnetic excitation in reciprocal space is to be followed to prove its nature. The results will be discussed subsequently in the frame of other vdW materials.

### Results:

The vanadium trihalide VBr<sub>3</sub> has been synthesised in the form of single crystals. Six single crystals were co-aligned and glued using GE-varnish to two aluminium plates to maximise the sample volume in the beam. The sample was aligned using the hexagonal description of R-3 structure (space group n. 148) with the hh-plane to be the scattering plane. Only several nuclear reflections with non-zero intensity were available employing the spectrometer setup with  $k_f = 1.5 \text{ \AA}^{-1}$ . (We used reflections (110), (-210), and (300) for sample alignment.) The expected change in crystal structure below 90 K (as deduced from magnetisation and specific heat data and reports on other trihalides) was not observed on sample cooling, most likely due to mosaicity of the crystal alignment and instrument resolution. Therefore, a hexagonal description of the lattice was used down to a base temperature of 1.6 K.

Based on our bulk properties data and first-principle calculations, we expected to observe magnetic signal on top of nuclear reflections. The (000) propagation vector was not confirmed by our INS experiment on ThALES. No clear additional magnetic signal on nuclear reflections was observed below ordering temperature. Nevertheless, a clear magnetic signal evolving with temperature was followed in non-zero energy transfer data (Fig.1). A critical scattering behaviour was followed at selected Q-position (-110) at 1 meV energy transfer.

Most of the experimental time (5 days) was dedicated to measurements of magnetic excitations. First, several long Q-scans were measured at constant energy transfer of 2.5 meV. See the diagram in Fig.2 for Q-directions of measurement (1 day). Subsequently,  $\Delta E$ -scans were measured following these Q-directions. The example in Fig.3 shows an evolution of magnetic excitation with Q up to higher energies.

The performed experiment showed the feasibility of neutron scattering investigation of  $VBr_3$  trihallide, as well as the good bulk-quality of our single crystals. Critical magnetic scattering was observed, pointing out strong short-range correlations above magnetic ordering temperature. Magnetic excitations were observed and their evolution with energy and  $Q$  was followed. However, several ingredients to fully understand the system. First, knowledge on magnetic propagation vector and magnetic structure is essential for the correct interpretation of measured data. Second, an inelastic neutron scattering experiment in the  $h$  $l$ -plane (to complement the current  $h$  $h$ -plane experiment) is highly desirable. Third, measurement up to higher energy transfers seems to be compulsory to unambiguously determine the dispersion relations of observed magnetic excitations. We intend to address these by proposing further elastic and inelastic neutron experiments in the near future.

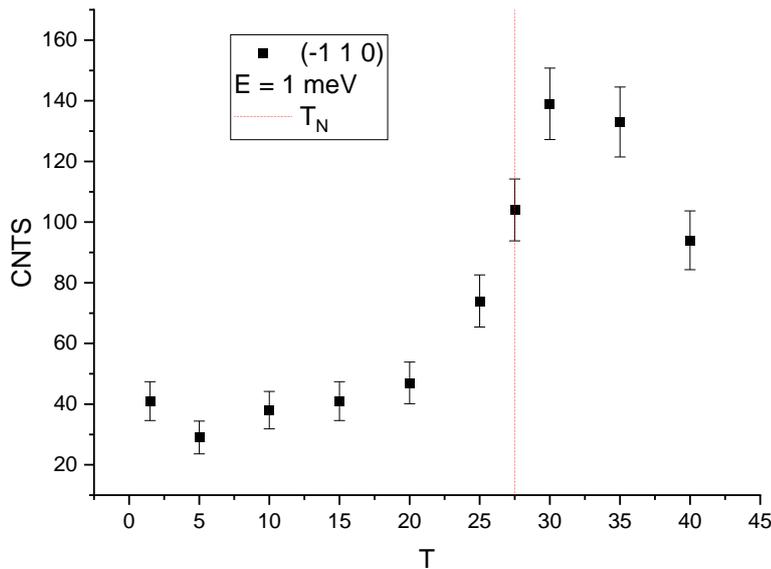


Fig.1 – Critical behaviour in  $VBr_3$ . The temperature dependence of the magnetic signal is followed at  $Q = (-110)$  and  $\Delta E = 1$  meV. Ordering temperature determined from magnetisation measurements is depicted as well.

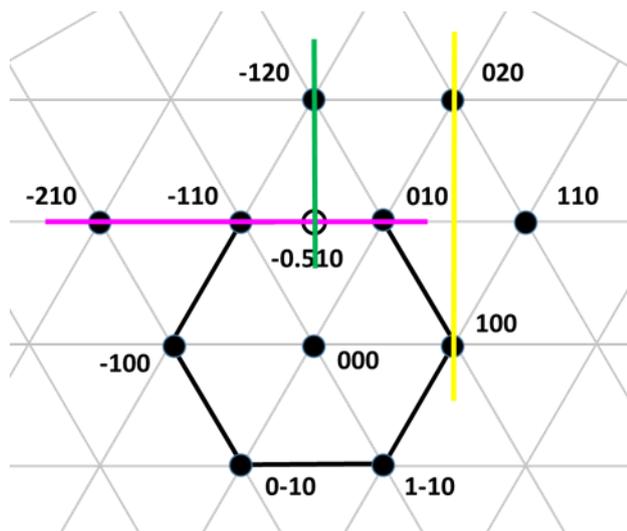


Fig.2 –  $h$  $h$ -plane of the hexagonal lattice. Reflections and  $Q$ -directions (coloured lines) mapped within the current experiment are depicted.

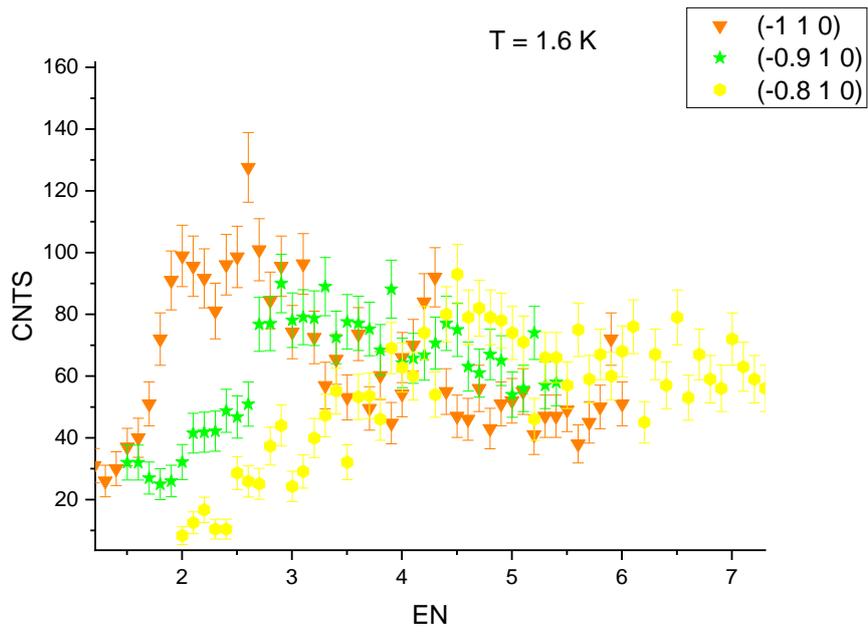


Fig.3 – Examples of measured magnetic excitations in VBr<sub>3</sub>. Excitations shift to higher energy with Q decreasing (increasing) from (-110) position.