

# Experimental report

12/11/2018

**Proposal:** 4-05-705

**Council:** 4/2018

**Title:** Magnetic excitations of a new potential spin liquid

**Research area:** Physics

**This proposal is a new proposal**

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**Local contacts:** Jacques OLLIVIER

**Samples:** CuSb<sub>2</sub>O<sub>6</sub>

Instrument	Requested days	Allocated days	From	To
IN5	3	1	12/09/2018	13/09/2018

## Abstract:

We have synthesized CuSb<sub>2</sub>O<sub>6</sub> in a new structure, the rosiaite (PbSb<sub>2</sub>O<sub>6</sub>) structure, space group P-31m. In this structure magnetic cations are arranged in trigonal layers, and the oxygen octahedron around the copper is more symmetric than in the common [slightly distorted] trirutile structure. The absence of long-range order and of ZFC-FC splitting in the susceptibility, the isotropy of the Cu<sup>2+</sup>, and the presence of sizable antiferromagnetic interactions (deviations from Curie-Weiss below ~150K) could imply a quantum (S=1/2) spin liquid scenario, expected for triangular antiferromagnets with nn/nnn interactions.

We propose to investigate the magnetic excitations of CuSb<sub>2</sub>O<sub>6</sub> on IN5 at 5 temperatures, and ask for 3d of beamtime. We need IN5 for its superior resolution and flexibility.

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## Scientific context

$\text{MSb}_2\text{O}_6$  ( $\text{M}=\text{Co}, \text{Ni}, \text{Cu}, \text{Zn}$  and  $\text{Mg}$ ) usually crystallize in the tetragonal trirutile form [1],  $\text{CuSb}_2\text{O}_6$  slightly distorts into a monoclinic structure due to the Jahn-Teller effect. We have synthesized  $\text{MSb}_2\text{O}_6$  in a new structure, the rosielite ( $\text{PbSb}_2\text{O}_6$ ) structure, space group  $\text{P-31m}$ . In this structure magnetic cations are arranged in trigonal layers. In  $\text{M}=\text{Co}$  and  $\text{Ni}$  these layers antiferromagnetically order at low temperatures (11K and 15K respectively), forming spin-frustrated triangles. Neutron diffraction studies confirm the rosielite-type structure for  $\text{CuSb}_2\text{O}_6$  as well (magnetic cations  $\text{Cu}^{2+}$  [2]). Lattice constants of  $\text{CuSb}_2\text{O}_6$  are  $a=b=5.054(4)$  Å,  $c=4.5881(10)$  Å [2]. The triangular antiferromagnetic (AF) Heisenberg model is a typical example of two-dimensional geometrically frustrated magnets. With only AF nearest-neighbor interaction the ground state of this system is the three-sublattice  $120^\circ$  structure, which is commensurate to the underlying lattice. With further-neighbor interactions spin liquid or skyrmion phases can be realized [3-5]. The absence of long-range order, the isotropy of the  $\text{Cu}^{2+}$ , the presence of sizable antiferromagnetic interactions in  $\text{CuSb}_2\text{O}_6$  [2] could imply a quantum ( $S=1/2$ ) spin liquid scenario.

## Experiment details

We performed an experiment on IN5 from the 12th to 14th of September 2018 with the aim to identify the character of the magnetic excitations. We used Orange Cryostat reaching a base temperature of 1.5K. We used  $\lambda=2.6$  Å and 4.8 Å. We had a powder sample of 10.5g in the hollow cylinder. Additionally, the neutron diffraction experiment EASY-363 at the room temperature was performed in order to verify the crystal structure of this compound (see experimental report).

## Conclusion

With  $\lambda=2.6$  Å, we have performed measurements at the base temperature 1.6K and at the 50K – below and above the temperature where the deviation of the susceptibility from the Curie-Weiss law is visible. At 1.6K we can see excitations emerging at about  $0.68$  Å $^{-1}$  with a steep dispersion that reaches out to about 5-6 meV. They are absent at high temperature (Fig.1 right shows 50K data) and therefore most likely of magnetic origin.

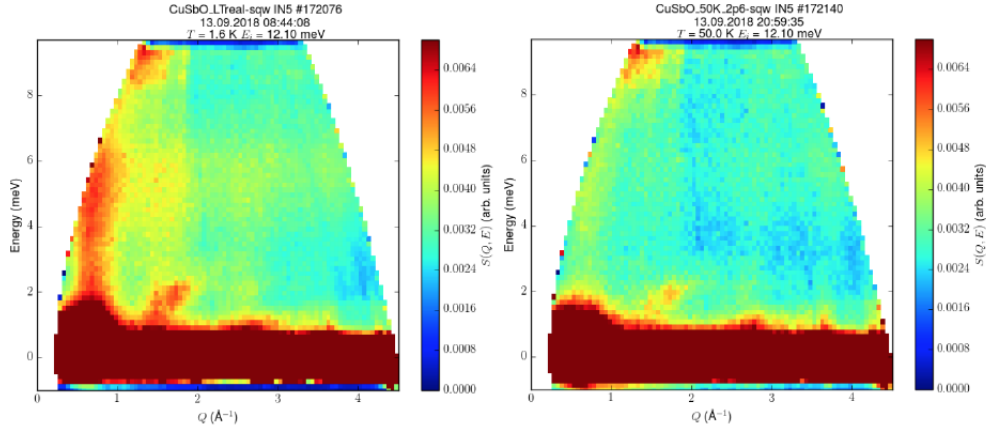


Figure 1: Low-Q magnetic excitations (left) at 1.6K and its absence (right) at 50K with  $\lambda=2.6$  Å. The "dispersive feature" near  $1.5$  Å $^{-1}$  is spurious, it is absent at other incident energies.

After subtracting an empty can spectrum at the base temperature the spectrum looks more clear and the intensive feature at the very high energies is gone (Fig.2).

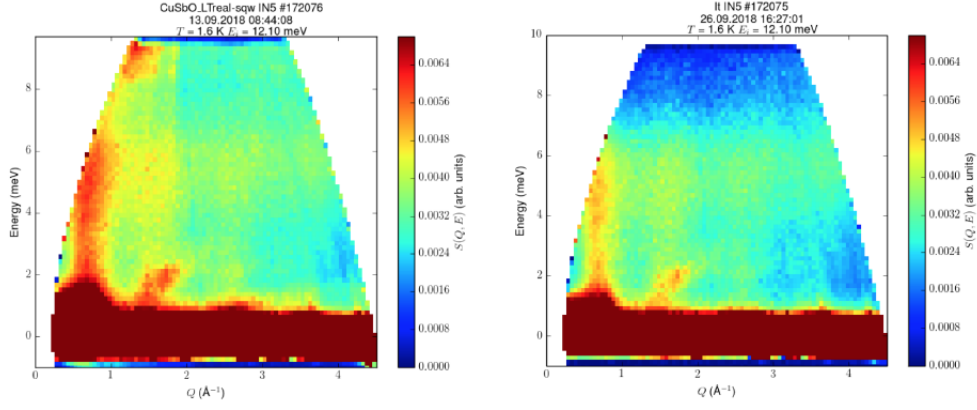


Figure 2: Low-Q magnetic excitations at 1.6K without (left) and with (right) subtraction of the empty container.

With wavelength  $4.8 \text{ \AA}$  we did not detect any gap down to about  $0.2 \text{ meV}$  near  $0.68 \text{ \AA}^{-1}$ . An additional lobe of low-lying excitations is visible near  $1.6 \text{ \AA}^{-1}$ , it reaches out to maximum  $1 \text{ meV}$  and disappears already at  $10 \text{ K}$  (Fig. 3).

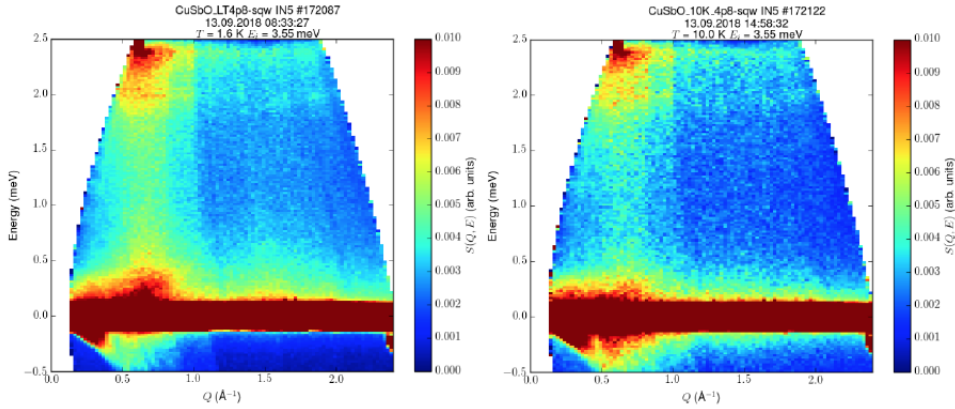


Figure 3: Spectra of CuSb<sub>2</sub>O<sub>6</sub> with  $\lambda=4.8 \text{ \AA}$  (left  $T=1.6 \text{ K}$ , right  $T=10 \text{ K}$ ). The feature at low  $Q$  and energy transfer  $2\text{-}2.5 \text{ meV}$  is of spurious origin, it is absent at other incident energies. The feature at very low  $Q$  and negative energy transfer is likewise spurious.

After subtracting an empty can spectrum at the base temperature the spectrum looks more clear and the intensive feature at the very high energies as well as the negative energy transfer feature are gone (Fig.4). Overall, the diffuse character of the observed excitations is very similar to the dynamic response in 2D quantum spin liquids [6]. A gapless spin-liquid with algebraically decaying correlations is predicted for the triangular J1-J2 Heisenberg antiferromagnet for a range of interactions  $J2/J1$  between about  $0.05$  and  $0.17$  [4], sandwiched between the  $120$  degree Neel state and the collinear striped AF ground state. Of course, a deep analysis of the obtained IN5 data need to be done, but in order to verify that we are really dealing with an isotropic quantum spin liquid displaying gapless excitations, we would like to confirm the absence of an anisotropy gap at low energies at dilution temperatures.

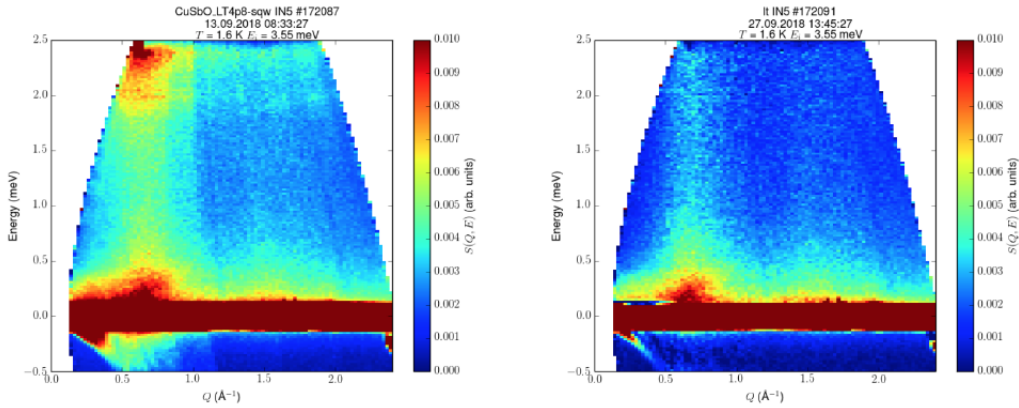


Figure 4: Spectra of CuSb<sub>2</sub>O<sub>6</sub> with  $\lambda=4.8 \text{ \AA}$  at  $1.6 \text{ K}$  without (left) and with (right) subtraction of the empty container.

## References

- [1] W. H. Baur, Crystallography Reviews 13, 65 (2007).
- [2] A. Y. Nikulin et al, Dalton Transactions 46, 6059 (2017).
- [3] T. Okubo et al, Phys. Rev. Lett. 108, 017206 (2012).
- [4] P. Li et al, Phys. Rev. B 91, 014426 (2015).
- [5] H. D. Rosales et al, Phys. Rev. B 92, 214439 (2015).
- [6] B. Fak et al, Phys. Rev. Lett. 109, 037208 (2012); B. Fak et al, Phys. Rev. B 95, 060402(R) (2017).