Experimental report

12/11/2018

Proposal: **4-01-1574** **Council:** 4/2017

Title: Magnetic excitations of a spin tube

Research area: Physics

This proposal is a new proposal

Main proposer: **Mechthild ENDERLE**

Experimental team:

Local contacts: Mechthild ENDERLE
Martin BOEHM

Samples: CuSiO3.H2O

Instrument	Requested days	Allocated days	From	To
THALES	7	7	22/03/2018	29/03/2018
IN20 Flatcone	4	4		

Abstract:

The rhombohedral gem-stone mineral green dioptase CuSiO3„eH2O contains Cu2+-oxygen spirals along the hexagonal c-axis with a honeycomb arrangement of the spirals in the ab-plane. Susceptibility, specific heat, neutron diffraction, and quantum chemical calculations of the exchange paths point to a quasi-one dimensional Heisenberg antiferromagnetic chain behaviour with antiferromagnetic intrachain exchange of 74K, and ferromagnetic interchain interaction of 6K. However, susceptibility and Raman data have been interpreted as evidence for dimer formation in the ab-plane, with a dimer gap of 6meV and a two-dimer excitation of 10.5 meV. This scenario locates green dioptase close to a quantum critical point. We propose to study the magnetic excitation spectrum of green dioptase, in order to establish a reliable scheme of exchange integrals. We wish to perform overview measurements on IN20 and Thales using FlatCone, completed by well-resolved single-detector scans on Thales.

Experimental report 4-01-1574 – Magnetic excitations of a spin tube

I. Safiulina, M. Enderle, M. Boehm

Scientific context

The gem-stone mineral green dioptase with the chemical composition $CuSiO_3Si{\cdot}H_2O$ is built by hexagonal rings of silica tetrahedra, $(Si6O18)^{12-}$, interconnected by the magnetic Cu^{2+} ions with spin 1/2. It crystallizes in the space group R-3. The Cu^{2+} are surrounded by axially-elongated oxygen octahedra [1], [2]. The copper-oxygen network forms cornersharing spirals along the hexagonal c-axis, neighbouring copper sites along the spiral are displaced by c/3. The spiral chains have a honeycomb arrangement in the ab-plane (Fig. 1).

Susceptibility [3], [4], specific heat [5], [6], neutron diffraction [2], NMR [7] and quantum chemical calculations of the exchange paths [6] point to an unfrustrated quasi-one dimansional Heisenberg antiferromagnetic chain behaviour with antiferromagnetic intrachain exchange of 74K, and smaller ferromagnetic interchain coupling. Specific heat measurements [5], [6] evidence the onset of long-range order with an extremely sharp λ-anomaly at T_N=14.5K.

In short, the theoretical description of the material is contradictory [1-10], while the experimental findings so far indicate a material that appears dominated by quantum correlations.

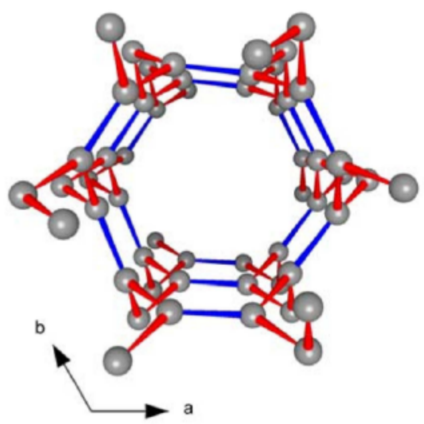

Figure 1: Central projection of the hexagonal basal plane of green dioptase along c (only Cu atoms are shown). The connectivity of the Cu atoms in the spiral chains (intra-chain) are highlighted in red, the dimer resp. ferromagnetic interchain bonds are highlighted in blue [1], [2].

Experiment details

We performed polarized inelastic experiment on Thales from the 22nd to 29th of March 2018 with the aim to identify the energy range of the magnetic excitations. The polarisation was varying by the Helmholtz coils.

Conclusion

Before the experiment we have performed spin-wave calculations for two different cases - the magnetic moment pointed along the c-axis [11] and the moment inclined from the c-axis by 13 degrees [2] (Fig.2).

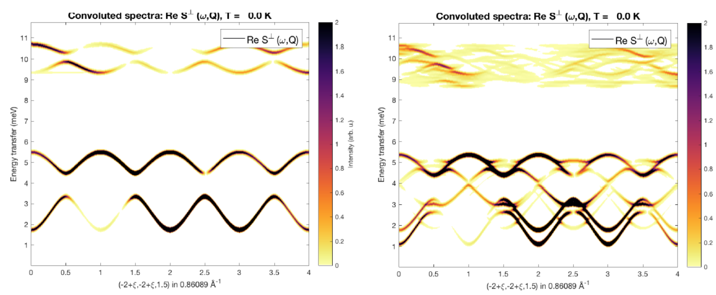

Figure 2: Left: spin-wave spectrum for the magnetic moment pointed along c-axis; Right: spin-wave spectrum for the magnetic moment inclined by 13 degrees off the c-axis.

As you can see more modes for the second case are expected. So that, at the certain Q-values we have measured inelastic spectra to understand the nature of such excitations (Fig.3).

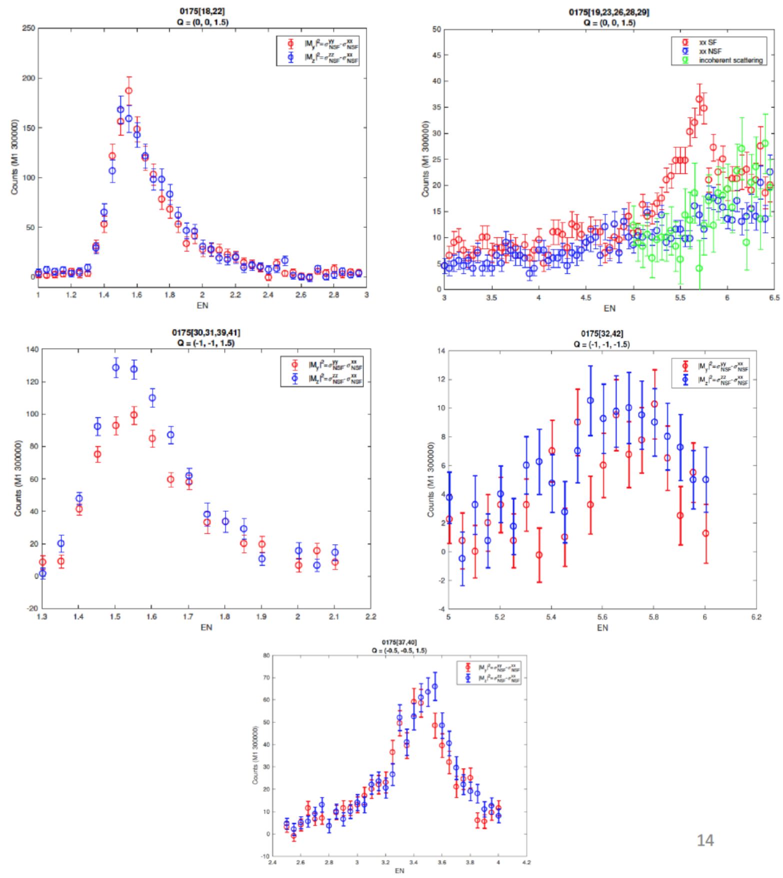

Figure 3: Examples of the inelastic spectra for different Q-values.

At the obtained spectra you can always see a "long tail" that can be because of two reasons. First, such a long tail could be a result of containing several peaks in it what will lead to the inclination of the magnetic moment as it shown on the Fig.2 right. Second, this could be because of "continuum" which is related to the quantum nature of spin-1/2 Cu-ions and not fully ordered moment.

To understand the nature of such excitations first we need to define the magnetic structure, namely, the direction of the magnetic moment with the D3 experiment which has already been performed (prop. N 5-54-252). Deep analysis of the D3 experiment will allow us to see the possible inclination of the magnetic moment and then with this knowledge we will perform spin-wave calculations again and will directly compare it with the Thales results.

References

[1] W. Eysel and K.-H. Breuer, Z. Deutsch. Gemmol. Ges. 30, 219 (1981).
[2] E. L. Belokoneva et al., Phys. Chem. Minerals 29, 430 (2002).
[3] R. E. Newnham and R. P. Santoro, Phys. Stat. Sol. 19, K87 (1967).
[4] M. Wintenberger et al., Solid State Comm. 87, 309 (1993).
[5] I. A. Kiseleva et al., J. Chem. Thermodynamics 25, 621 (1993).
[6] J. M. Law et al., Z. Anorg. Allg. Chem. 636, 54 (2010).
[7] K. Matsui et al., JPS Conf. Proc. 3, 014011 (2014).
[8] C. Gros et al., Europhys. Lett. 60, 276 (2002).
[9] O. Janson et al., Phys. Rev. B 82, 014424 (2010).
[10] H. Ohta et al., J.Phys. Conf. Ser. 150, 042151 (2009).
[11] A. Podlesnyak et al., Phys. Rev. B 93 (2016).