Experimental report

12/11/2018

Proposal: 5-54-252 **Council:** 4/2017

Title: Magnetic structure of a spin tube

Research area: Physics

This proposal is a new proposal

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Samples: CuSiO3.H2O

Instrument	Requested days	Allocated days	From	To
D3 CPA	7	6	20/04/2018	21/04/2018
			04/09/2018	09/09/2018
ORIENTEXPRESS	1	1	16/04/2018	17/04/2018

Abstract:

The rhombohedral gem-stone mineral green dioptase CuSiO3.H2O 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. A low ordered moment and a large spin-flop transition field of 13.5T are difficult to reconcile.
We propose to reinvestigate the magnetic structure of green dioptase using single-crystal polarised neutron diffraction on D3.

Experimental report 5-54-252 – Magnetic structure of a spin tube

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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.

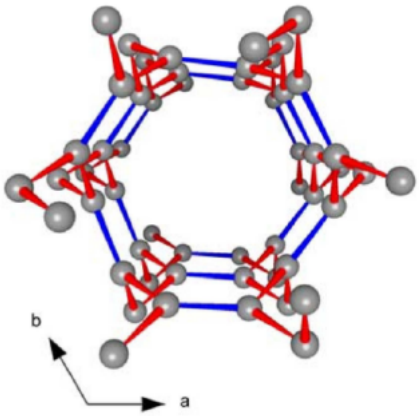

Figure 1: Central projection of the hexagonal basal plane of green dioptase along c (only Cu atoms are shown). The connectivity of the Cu atoms in the spiral chains (intra-chain) are highlighted in red, the dimer resp. ferromagnetic interchain bonds are highlighted in blue [1], [2].

Experiment details

We performed polarized experiment on D3 from the 04th to 09th of September 2018 with the aim to determine the possible inclination of the magnetic moment or its absence due to the contradictory of several papers (for example, [2] and [11]). We used Cryocradle device with a base temperature of 1.5K in order to reach different hkl-planes and its polarisation tensors. We used two types of diaphragms in case to improve the data for some not highly intensive peaks. With λ=0.85 we have measured several Bragg reflections in different hkl-planes. It should be noticed that before this experiment we had a test experiment on IN20 TEST-2825 (also with the polarisation analysis).

Conclusion

To analyze the data (both from IN20 and D3) we used Mag2Pol program. We used first a model in which the moment is pointing along c-axis. From this, we could find the most intensive Bragg-reflections, and then, mostly important, the intensive reflections in which the polarisation elements (P_{yy}, P_{zz}, P_{yz}, P_{zy}) are the most sensitive to the inclination of the magnetic moment. From IN20 TAS experiment we've obtained inclinations of the moment about 11.5±0.8 degrees what is about E.Belokoneva's paper [2]. From D3 experiment we would like to obtain not only the inclination off the c-axis, but also its direction in the ab-plane, so, to know exactly the direction of the moment. After fitting D3 experiment we have obtained the same inclination as on IN20 (about 12 degrees) but the direction of the moment in the ab-plane for a moment is not so stable (Fig.2). For this purpose more detailed analysis need to be performed (especially for the peaks of not so high intensity). But the most important thing that we have observed the inclination of the moment and after full analysis of the D3 experiment we can easily analyze Thales inelastic data on this compound.

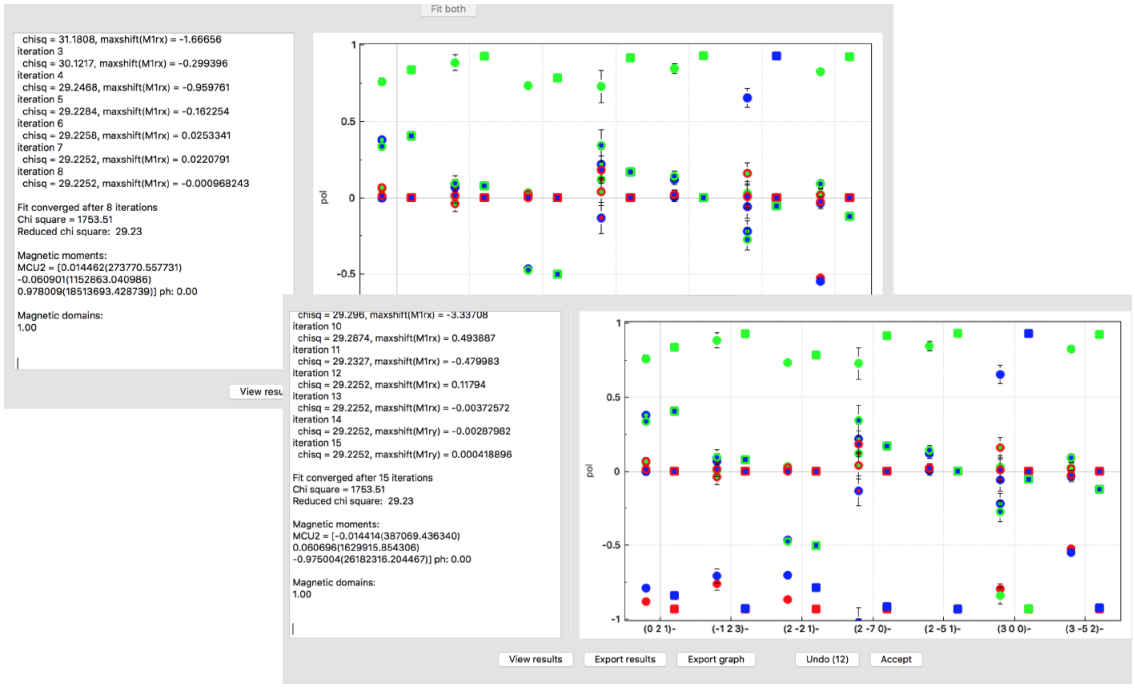

Figure 2: Example of the preliminary D3 fitting with opposite values of Mx and My - the components of the magnetic moment

References

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