Experimental report

13/05/2022

Proposal:	**5-32-923**	**Council:**	4/2021
Title:	Verifying the magnetic origin of the low-temperature diffuse signal in antlerite powder		
Research area:	Physics		

This proposal is a new proposal

Main proposer:	**Anton KULBAKOV**
Experimental team:	Andrew WILDES
Local contacts:	Andrew WILDES

Samples: Cu3SO4(OD)4

Instrument	Requested days	Allocated days	From	To
D7	3	5	18/06/2021	23/06/2021

Abstract:

The spin-1/2 low-dimensional quantum magnet antlerite, Cu3SO4(OH)4, is a natural mineral with a magnetic lattice consisting of three-leg ladders that are made of two nonequivalent Cu2+ ions with zigzag bonds. We synthesized large amounts of antlerite in the form of tiny single crystals, and we prepared much smaller deuterated crystals for the neutron diffraction experiment in order to determine the magnetic structure of the four different zero-field magnetic phases that were so far reported: ¿1, ¿2, I1 and I2. The main goal of the proposed experiment is to verify the magnetic origin of the observed diffuse signal, distinguish it from nonmagnetic background using polarized neutrons, and outline its region of presence in momentum space and in temperature

Experiment Title

Verifying the magnetic origin of the low-temperature diffuse signal in antlerite powder (#5-32-923)

Proposer

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Report

Experimental configuration and results

Our deuterated antlerite powder was measured on 18—23 June 2021, in vanadium can on D7 - diffuse scattering spectrometer at ILL, France. Neutron with the wavelength $\lambda = 4.8707$ Å were selected using 3 x HOPG (002), each with vertical focusing monochromator and polarized with Focusing bender supermirror (Co/Ti, m=2.8) polarizer. We are interested in temperature range 1.5—300 K, based on the recent magnetic phase diagram [1], and good temperature stability was crucial for our experiment since some phases are very narrow in temperature. Some of these data are already available in [2].

Figure 1 shows the diffuse magnetic intensity collected on the D7 diffractometer to higher temperatures. The data at all temperatures are fit to $I = AF^2$, where I is the intensity, F is the magnetic form factor for Cu^{2+}, and A is 0.2290(19) for all datasets in the units used. The diffuse peak shifts to lower $|\mathbf{Q}|$, but a significant diffuse contribution persists to at least 200 K. Spin-flip scattering was also performed to higher temperatures, as shown in Fig. 1, where the diffuse peak is seen to shift to lower $|\mathbf{Q}|$ on warming but does not vanish up to 200 K, indicating persistent short-range correlations. We note that this method does not differentiate between elastic and inelastic scattering, therefore we cannot distinguish static and dynamic correlations. A change in energy would also be seen as an apparent change in $|Q|$ if the fluctuations were dynamic.

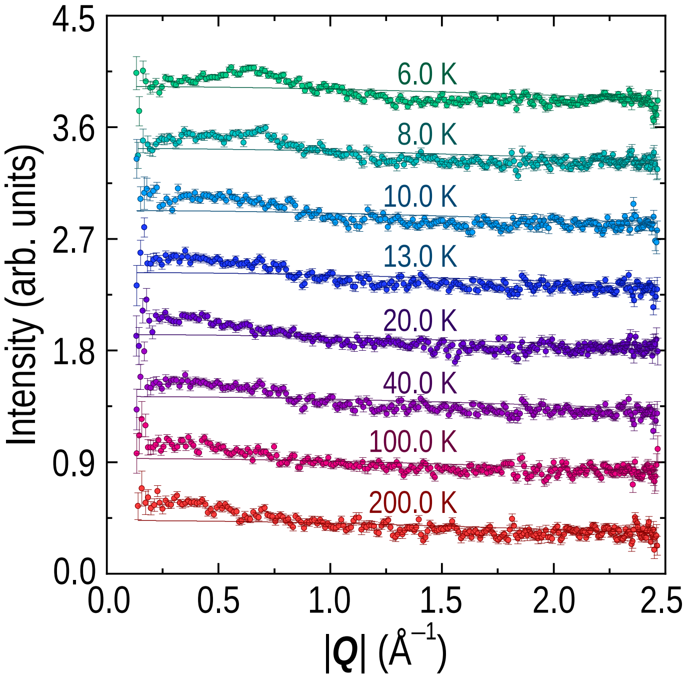

Fig. 1: Temperature evolution of magnetic signal. Solid lines represent the magnetic form factor of Cu^{2+}

We used the SPINVERT software package [3] to perform a reverse Monte Carlo refinement of the spin-flip neutron powder diffraction data, to recover the three-dimensional magnetic diffuse scattering pattern and the spin-pair correlation function. Reconstructed diffuse scattering intensity for antlerite in the paramagnetic phase at 6.0 K is shown in Fig. 2(b-d) for different cuts through reciprocal space. This is what we predict would be observed on a single crystal, if a sufficiently large crystal were available. Fig. 2(a) shows the resulting fit to the powder data, and Fig. 2(e) shows the extracted spin-spin correlation function. The shortest Cu–Cu distance in antlerite is a ferromagnetic exchange on the outer leg, followed by the antiferromagnetic link on the central leg and the second ferromagnetic exchange on the outer leg. Accordingly, the correlations in Fig. 2(e) are ferromagnetic at the shortest distance, then become mainly antiferromagnetic on average.

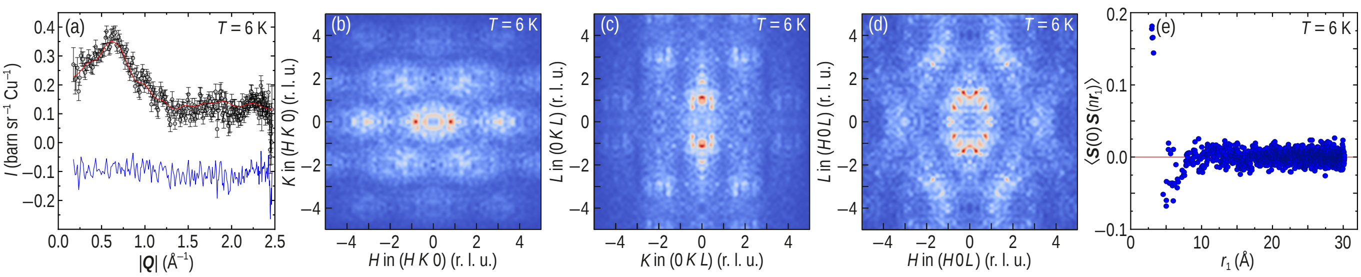

Fig. 2: Diffuse scattering in the paramagnetic phase at 6.0 K. (a) Fit (red) of the magnetic diffuse scattering intensity (gray), and its residual (blue). (b-d) Reverse-Monte-Carlo reconstructed scattering intensity for $(HK0)$, $(0KL)$ and $(H0L)$ cuts through reciprocal space, respectively. (e) Extracted spin-spin correlation functions as a function of distance. The ferromagnetic nearest neighbor correlations along the outer legs are visible at the lowest distances.

Beyond $\sim$8 Å the correlations average out to zero due to powder averaging — the sphere corresponding to this radius begins to include a large number of spins.

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[1] Y. Fujii *et al.*, J. Korean Phys. Soc. **62**, 2054-2058 (2013).
[2] A. Kulbakov et al., arXiv:2203.15343.
[3] J Paddison *et al.*, J. Phys.: Condens. Matter, **25**, 454220 (2013).