Experimental report

09/11/2018

Proposal: **INTER-421** **Council:** 4/2018

Title: Magnetic structure determinationof Ag2Cu2O3

Research area:

This proposal is a new proposal

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Experimental team: Clemens RITTER

Local contacts: Clemens RITTER

Samples: Ag2Cu2O3

Instrument	Requested days	Allocated days	From	To
D20	1	1	01/07/2018	02/07/2018

Abstract:

Experimental Report IN-421

Magnetic Structure Determination of the linear chain quantum antiferromagnet $Ag_2Cu_2O_3$

Cu_4O_3 ('paramelaconite') is a binary copper oxide which is chemically intermediate between Cu_2O ('cuprite')) and CuO ('tenorite'). The crystal structure contains interpenetrating rods of (diamagnetic) Cu^+ cations forming two collinear bonds with oxygen atoms and (magnetic, S=1/2) Cu^{2+} atoms forming four bonds to oxygen atoms at the corners of a rectangle .[1] The magnetic Cu^{2+} cations form tetrahedra similar to the arrangement in the cubic pyrochlores implying that geometrical frustration plays an essential role for the magnetic properties.[1,2] Preparation of bulk samples (neither powder nor x-tals) of Cu_4O_3 has not been feasible, so far. The magnetic properties (magnetic susceptibility and x-tal neutron diffraction) have therefore been done on a small piece of a natural Cu_4O_3 crystal. The magnetic susceptibility of Cu_4O_3 exhibits typical features of a low dimensional quantum antiferromagnet (afm) with a broad maximum at ~75 K and long-range afm ordering below ~40 K. The magnetic structure determined on the basis of a doubling of the tetragonal unit cell (SG $I4_1/amd$) in all spatial directions is a commensurate helical-type arrangement of ordered moments of 0.46 μ_B/Cu.[3] A collinear structure gives slightly worse reliability factors. However, published theoretical estimates of the exchange parameters could not explain the magnetic structure of Cu_4O_3.[4]

In contrast to Cu_4O_3, polycrystalline samples of the isotypic $Ag_2Cu_2O_3$ where the Ag^+ cations replace the Cu^+ cations can be prepared in larger quantities. $Ag_2Cu_2O_3$ exhibits long-range afm order below $T_{Néel}$ ~65 K (see Fig. 1a) and similar low-dimensional features as Cu_4O_3.[5,6] From x-ray powder diffraction experiments Uematsu *et al.* conjectured a distortion to a monoclinic structure (SGR $C2/c$) being present already at room temperature and showing a stark increase at $T_{Néel}$.[7]

The experiment on D20 was aimed to determine the magnetic the magnetic structure. We have collected three datasets at 1.5 K, 50 K and 80 K. The datasets at 50 K and 80 K were merged and taken as non-magnetic reference. The datasets were collected for 10 h with an instrument setting giving maximum flux at a wavelength of 2.41 A. In order to obtain lattice parameters at low temperatures a Rietveld refinement of the diffraction collected at 1.5 K with the atom positional parameters fixed to the values we have obtained from a preceding high-resolution experiment on MLZ's powder diffractometer SPODI (see Figure 1).

Using these lattice parameters we indexed Bragg peaks in the difference pattern (1.5 K minus 50+80 K) and derived at a tentative magnetic propagation vector of (0.5, 0.5 ~0.31) indicating an incommensurate magnetic structure perpendicular to the ribbon chains (c-axis in I 1 1 2/b, spgr no. 15). Figure 2 displays the Rietveld refinement of the difference pattern indicating magnetic moments of ~0.5μ_B and ~0.8 μ_B for the Cu atoms.

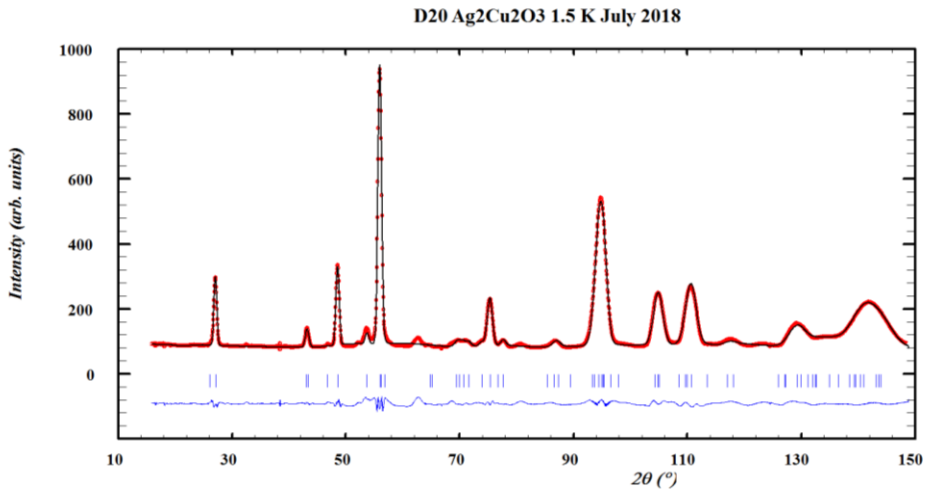

Fig. 1 Rietveld refinement of the neutron powder diffraction pattern collected at 1.5 K at D20 (λ = 2.41 Å).

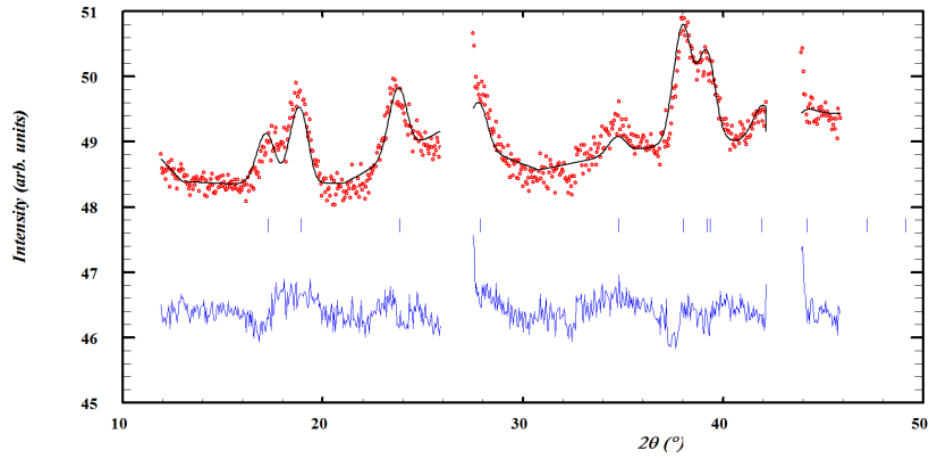

Fig. 2 (red circles) Difference of two NPD patterns of $Ag_2Cu_2O_3$ collected at 1.5 K and 80 K on D20 for 10h each. (black solid line) Tentative refinement of the difference pattern assuming a magnetic propagation vector of (½,½,0.31(1)). Vertical bars mark the positions of the magnetic Bragg reflections.

In a forthcoming proposal we plan to increase the statistics of our data by significantly enlarging the sample amount (currently ~8000 mg) and possibly increasing the counting time. The current sample was used because it had structurally been characterized in detail earlier from SPODI data.

References:

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[3] L. Pinsard-Gaudart, J. Rodríguez-Carvajal, et al., Phys. Rev. B **69**, 104408 (2004).
[4] M. H. Whangbo and H. J. Koo, Inorg. Chem. **41**, 3570 (2002).
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[6] K. Adelsberger, J. Curda, S. Vensky, and M. Jansen, J.Solid State Chem. **158**, 82 (2001).
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