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Elastic neutron diffraction study of long-range antiferro-magnetic order in the $S = \frac{1}{2}$ quantum chain system $CuSb_2O_6$

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Abstract

$CuSb_2O_6$ crystallizes with a monoclinically distorted variant of the trirutile structure type with Cu^{2+} ions arranged in square planar nets being structurally well separated from each other by Sb–O building blocks. Using elastic neutron diffraction we have established the low-temperature nuclear structure and antiferromagnetic (afm) arrangement of Cu moments at 2 K. The magnetic unit cell is doubled with respect to the nuclear cell along the *a*- and *c*-axes. The magnetic anisotropy in the ordered state suggests that Cu moments of magnitude 0.51(2) μ_B lie in the $a-b$ plane.

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Keywords: $CuSb_2O_6$; Uniform Heisenberg chain; Afm ordering; Elastic neutron scattering; Quantum-chain

1. Introduction

Efforts to understand the underlying microscopic mechanism of high T_C superconductivity have refocused attention on the magnetic behavior of quantum spin $S = \frac{1}{2}$ chain and ladder systems. The system $CuSb_2O_6$ crystallizes in a monoclinically distorted trirutile structure type. It has been reported to exhibit magnetic strong short-range order (SRO) signalled by a broad susceptibility maximum centered around 60 K [1]. The susceptibility was reported to fit reasonably well to an afm uniform $S = \frac{1}{2}$ Heisenberg chain ($J = -49.6$ K) model with nearest neighbor coupling. A sharp decrease in the susceptibility below 8.6 K indicates the onset of long-range afm order due to interchain interactions. Initial powder neutron diffraction suggested a propagation vector $k = (\frac{1}{2}, 0, \frac{1}{2})$ for the afm ordered phase. Various ordering models have been discussed in Ref. [2].

2. Experimental

Single crystals were grown by chemical vapor transport as described in detail previously [3]. Magnetic susceptibilities were measured in a MPMS7 magnetometer. Using the four circle diffractometer D10 (ILL, Grenoble) a single-crystal neutron diffraction measurement was carried out. The structure refinement was performed with standard routines of the Cambridge Crystallographic Subroutine Library [4].

3. Results and discussion

Fig. 1 displays the magnetic susceptibility and the heat capacity of a crystal of $CuSb_2O_6$ [4]. Long-rang order is evident from the heat capacity anomaly and the kinks in

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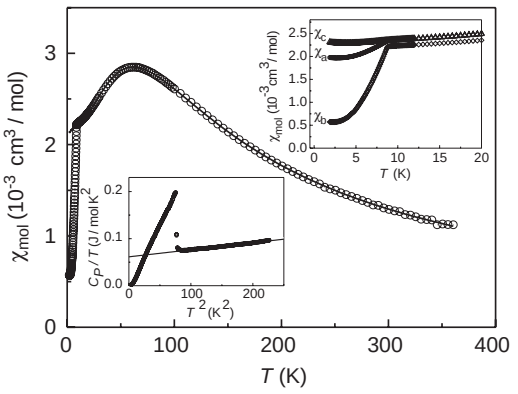

Fig. 1. Susceptibility of a single crystal of $CuSb_2O_6$ measured in a field of 0.1 T along the b-axis (B exp., full line: fit, see text). (a) Heat capacity indicating long-range ordering at 8.68(5) K. (b) Susceptibility along the c-, a-, and b-axis as indicated (top to bottom).

the susceptibility below $T_N = 8.68(5)$ K. The fit of the susceptibility with the model of an afm $S = \frac{1}{2}$ Heisenberg chain with uniform nearest-neighbour coupling yields close to perfect agreement ($|\chi^{fit}_{mol} - \chi^{exp}_{mol}| \leqslant 0.4\%$; $10\,K < T < 350\,K$) using an exchange parameter of $J = -48.0(5)$ K. In addition to a phonon term ($\propto T^3$), the low-temperature heat capacity ($10\,K < T$) contains a linear magnetic term ($\propto T$) consistent with the expected value $C_{mag}/R = T/3J$ for a uniform afm $S = \frac{1}{2}$ Heisenberg chain [3,5].

Below T_N the susceptibility exhibits pronounced anisotropy. With the field aligned along the a- or b-axis, χ drops while for the field along the c-axis no decrease below T_N can be detected. This observation suggests that the ordered moment lie in the $a-b$ plane with their major component along b.

Single crystal neutron diffraction on $CuSb_2O_6$ at room temperature and at low temperatures revealed additional weak Bragg reflections below T_N which can be indexed on the basis of a propagation vector $k = (\frac{1}{2}, 0, \frac{1}{2})$. A refinement of the nuclear structure at low temperature gave no indication of a structural phase transition. Using two different magnetic structure models and 89 magnetic reflections the refinement of the magnetic structure based on 89 magnetic reflections converged well with identical χ^2 and $R_2 = 7.4\%$ and 7.7% with moments either in the $a-b$ or the $b-c$ plane, respectively.[1] For both models the ordered moment at 2 K amounts to $0.51(2)\,\mu_B$ in good agreement with the estimate given in Ref. [2]. On the basis of the anisotropy of the susceptibility we suggest a magnetic structure as displayed in Fig. 2 with moments tilted 103(6)° (Cu1: (0,0,0)) and 70(5)° (Cu2 : $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$) away from the a-axis and oriented antiparallel along (1 1 0) [4].

[1] R_2 is defined as the ratio of the sum of the weighted differences (observed−calculated) and the sum of the weighted observed intensities.

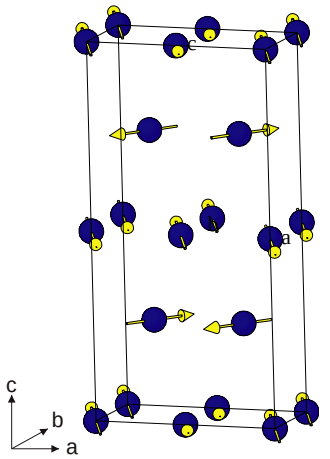

Fig. 2. Arrangement of the ordered Cu^{2+} moments in the afm long-range ordered phase of $CuSb_2O_6$.

4. Summary

Magnetic susceptibility, heat capacity and neutron diffraction of a single crystal of $CuSb_2O_6$ indicate $S = \frac{1}{2}$ Heisenberg chain behavior above and long-range afm ordering below $T_N \approx 8.7$ K. The magnitude of the ordered Cu^{2+} moment amounts to $0.51\,\mu_B$ and they lie in the $a-b$-plane with the major component along the b-axis.

5. Note added in proof

While proofreading we became aware of a neutron diffraction study on $CuSb_2O_6$ by Kato et al. [J. Phys. Soc. Jpn. 71 (2002) Suppl. 187 (2002)].

Acknowledgements

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