Experimental report

13/04/2026

Proposal: DIR-320 **Council:** 10/2023

Title: Unravelling the ground state magnetic order of a Kagome-like lattice

Research area: Physics

This proposal is a new proposal

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Samples: Cu2OSO4

Instrument	Requested days	Allocated days	From	To
D3 High field >1T	5	5	12/06/2024	17/06/2024
D9	5	5	11/04/2024	16/04/2024
ORIENTEXPRESS	1	1	10/04/2024	11/04/2024

Abstract:

This proposal concerns the study of the ground state magnetic order of an unconventional Kagome lattice. These structures are known for their propensity towards alleviating the frustration of an otherwise ideal Kagome lattice, leading to the establishment of long-ranged magnetic order. One prime example is Cu2OSO4, in which one of the three corners of the conventional triangle is replaced by a dimer.

Our recent study of the spin dynamics of Cu2OSO4 on the CAMEA spectrometer (SINQ) point to a complex excitation spectrum dynamics, which cannot be captured by linear spin-wave theory along. Moreover, its unclear what the role of the dimer is. In order to develop a model Hamiltonian for Cu2OSO4, it is imperative that we determine the ground state magnetic order of the Cu sublattice, unambiguously.

To that end, we propose to measure the magnetic order and magnetization density of Cu2OSO4 using the 4-circle diffractometer D9 using the 2-circle half-polarised set up on D3, respectively. This combined D3 and D9 study will allow us to not know understand the spin configuration, but also the magnetization of the dimer in Cu2OSO4.

Di-copper oxygen sulphate (Cu_2OSO_4) is a realization of a kagome-like lattice of Cu ions, in which one of the three vertices of the conventional triangle is replaced by a dimer whose Cu atoms reside slightly out of the plane. The crystal structure therefore contains two inequivalent Cu sites: Cu1 ions (blue), which lie in the plane, and Cu2 ions (red), which form dimers [Fig. 1(a)]. As such, Cu_2OSO_4 can be regarded as a model system in which deviations from the ideal kagome lattice allow one to quantify the effects of fluctuations and strong frustration.

Previous powder and single-crystal neutron diffraction measurements enabled refinement of the magnetic ground state [1]. The Cu1 moments form non-collinear antiferromagnetic chains along the crystallographic *b* axis. These chains are connected by Cu2 dimers, which are ferromagnetically coupled. As a result, two Cu2 atoms and two Cu1 atoms form an approximately 120° spin arrangement involving four spins [Fig. 1(a)].

However, modelling inelastic neutron scattering data for this compound proved challenging. In addition, the single crystal previously used for diffraction exhibited significant mosaicity, with up to three distinct crystallites, preventing a systematic collection of Bragg peak intensities. For these reasons, we performed new measurements on a high-quality single crystal.

On D9, we first collected a dataset at 30 K, above the Néel temperature, to refine the nuclear structure and determine the extinction parameters of the crystal. We then collected a second dataset at 2 K, comprising 1142 reflections (586 independent), which enabled a full nuclear and magnetic refinement. The fit to the Bragg peak intensities is shown in Fig. 2(b). The refinement confirms a magnetic ground state consistent with previous results, indicating similar magnetic moment magnitudes on the two Cu sites and a relative angle of approximately 117.4°.

We subsequently performed measurements on D3 using a half-polarized setup to map the magnetization density on the copper sites. The same crystal used on D9 was cooled to 30 K and placed in a 9 T vertical magnetic field. Flipping ratios for a large number of Bragg reflections were collected, allowing reconstruction of the magnetization density using two approaches: the Fourier method [Fig. 2(a)] and the maximum entropy method [Fig. 2(b)]. Both methods consistently indicate a larger spin density on the Cu2 sites, elongated from the kagome plane toward the SO_4 groups. This observation suggests the importance of out-of-plane superexchange pathways. In contrast, the spin density on Cu1 ions appears weaker, despite the D9 refinement indicating comparable magnetic moment magnitudes on the two sites.

These findings, together with inelastic neutron data collected on CAMEA (SINQ) and IN12 (ILL), are helping to extrac the magnetic Hamiltonian for this system.

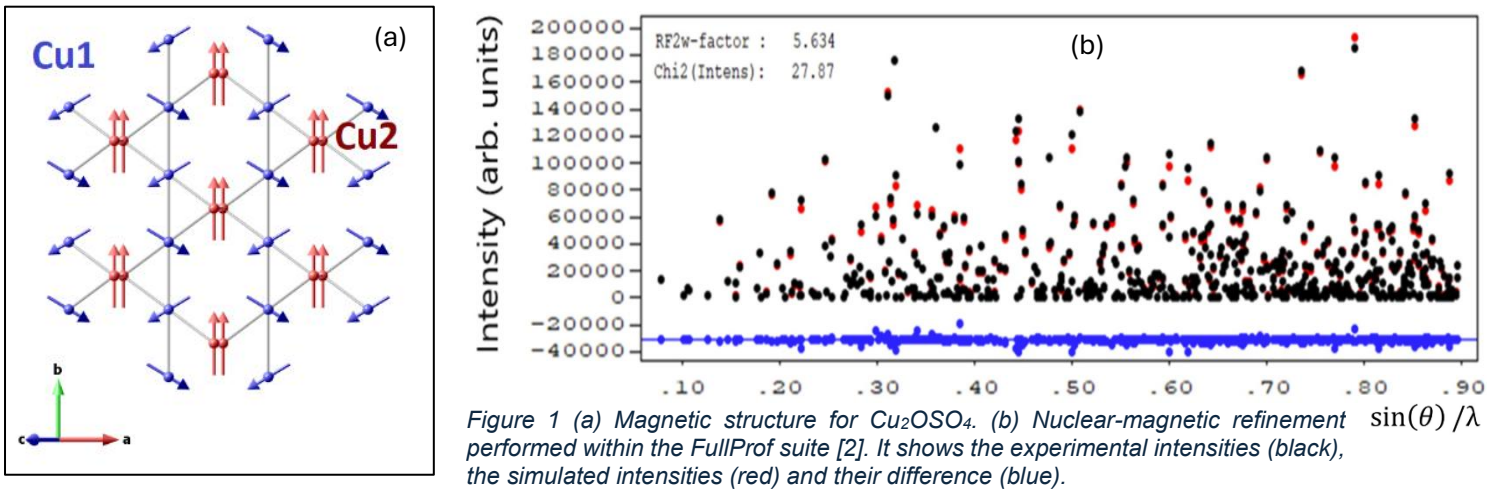


Figure 1 (a) Magnetic structure for Cu_2OSO_4. (b) Nuclear-magnetic refinement performed within the FullProf suite [2]. It shows the experimental intensities (black), the simulated intensities (red) and their difference (blue).

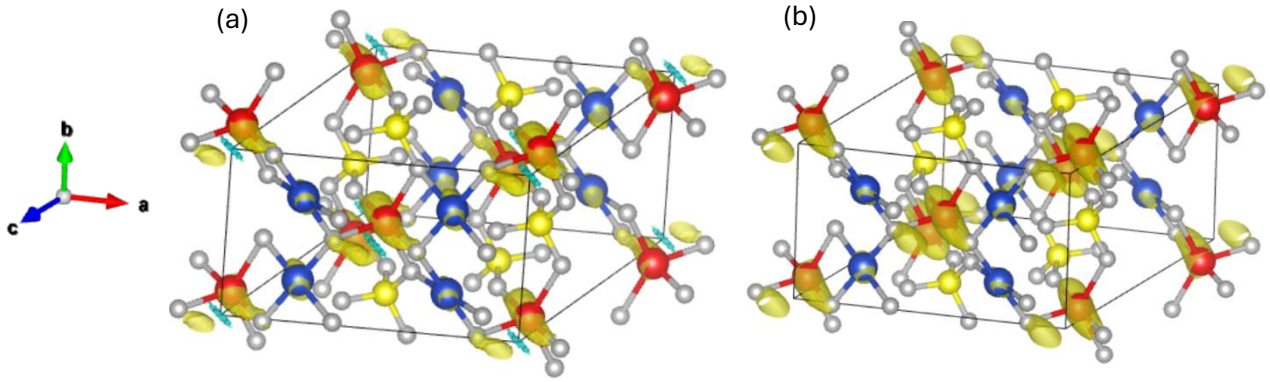


Figure 2 Magnetisation density reconstructed with the (a) Fourier and (b) maximum entropy methods. The atoms are Cu1 (blue), Cu2 (red), S (yellow) and O (grey).

References

[1] V. Favre et al., Physical Review B 102, 094422 (2020)

[2] Rodríguez-Carvajal, J., González-Platas, J., Katcho, Acta Cryst. B: Structural Science, 3, 302-317 (2025).