Experimental report

21/09/2026

Proposal: **5-31-2936** **Council:** 10/2022

Title: Noncollinear instabilities in the multiorbital magnet CuCr2S4

Research area: Physics

This proposal is a new proposal

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Samples: CuCr2S4

Instrument	Requested days	Allocated days	From	To
D20	2	1	03/07/2023	04/07/2023

Abstract:

In the cubic spinel compounds AB2X4 (X: chalcogen), the metal ions A and B form interpenetrating lattices of corner- and edge-sharing octahedra (A) and tetrahedra (B). The flexibility of substituting 3d transition metals opens up a fantastic potential for novel correlated electronic states defined by the competition of (in-)direct exchange paths. Using the local magnetic probes of muon spin rotation (¿SR) and Mössbauer spectroscopy, we have now uncovered the unexpectedly complex diagram in CuCr2S4. We propose a straightforward magnetic neutron powder diffraction experiment to clarify the low-temperature magnetism in CuCr2S4.

ILL EXPERIMENTAL REPORT

Noncollinear instabilities in the multiorbital magnet $CuCr_2S_4$

Proposal	5-31-2936
Instrument / dates	D20 / 3-4 July 2023
Experiment team	Vinicius Estevo Silva Frehse and Steven Gebel (on site); local contact: Clemens Ritter

Aim of the experiment

Following consultation with the local contact and the formal ILL sample-change procedure, we planned to measure $Fe_{1-x}Cu_xCr_2S_4$ (x=0.3, 0.7) instead of the originally proposed $CuCr_2S_4$. The Fe doped compound is also part of the project where we have been investigating the magnetic ground states of the $(Fe,Cu)Cr_2S_4$ metallic series. All compounds have the cubic spinel structure (AB_2X_4, *Fd-3m*), with the A and B sites featuring partially filled 3*d* orbitals.

Our preliminary characterization via Mössbauer spectroscopy and muon spin rotation (MuSR) suggested that (Fe,Cu) substitution induced a varied magnetic phase diagram [1], see Fig. 1(a). Specifically, based on the local magnetism alone, it is expected that crystals with Cu-concentration x=0.3 transition from a ferrimagnetic to a canted state at 100 K and then to a partially disordered magnetic (speromagnetic) state around 50 K. The x=0.7 samples, on the other hand, are expected to transition from ferrimagnetism to a distinct non-collinear phase with large net magnetization at 150 K, before entering a speromagnetic state around 50 K.

The aim of the experiment was to corroborate the interpretation of MuSR and Mössbauer data and identify the propagation vectors of potential magnetic superstructures that may be associated with the transitions.

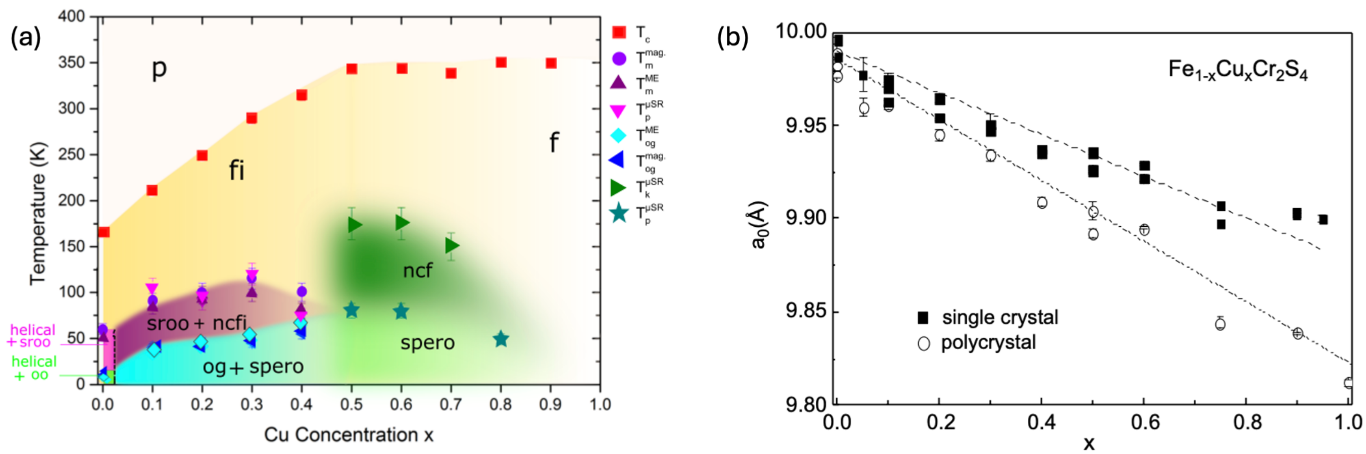

Figure 2. (a) Local-magnetism phase diagram of the $Fe_{1-x}Cu_xCr_2S_4$ series, based on MuSR and Mössbauer measurements, from [1]. (b) Characterization of previous batches using laboratory X-ray diffraction and electron-probe microanalysis showed homogeneous samples with lattice parameters conforming to Vegard's law [2].

Experiment

About 2 g each of polycrystalline $Fe_{1-x}Cu_xCr_2S_4$ with the nominal concentration (x=0.3, 0.7) were loaded into cylindrical vanadium cans. Trial measurements at λ = 1.54 and 2.41 Å established λ = 2.41 Å as the best wavelength to resolve peaks around similar positions. Final patterns were accumulated for 5 min between 1.5 K and 330 K with 2 K steps for the x=0.3 sample, and between 1.5 K and 360 K with 5 K steps for the x = 0.7 sample.

Results

The samples prepared for this beamtime had been screened by Cu Ka X-ray diffraction to confirm spinel phase purity. However, a weak splitting due to phase separation was overlooked. The refinement of the D20 data then revealed that the measured histograms are best modelled by proportional mixtures of the series' end compounds $FeCr_2S_4$ and $CuCr_2S_4$ (70%/30% and 30%/70% for x=0.3 and 0.7 respectively). As those compounds are part of a separate studies, the present experiment did not provide new insights.

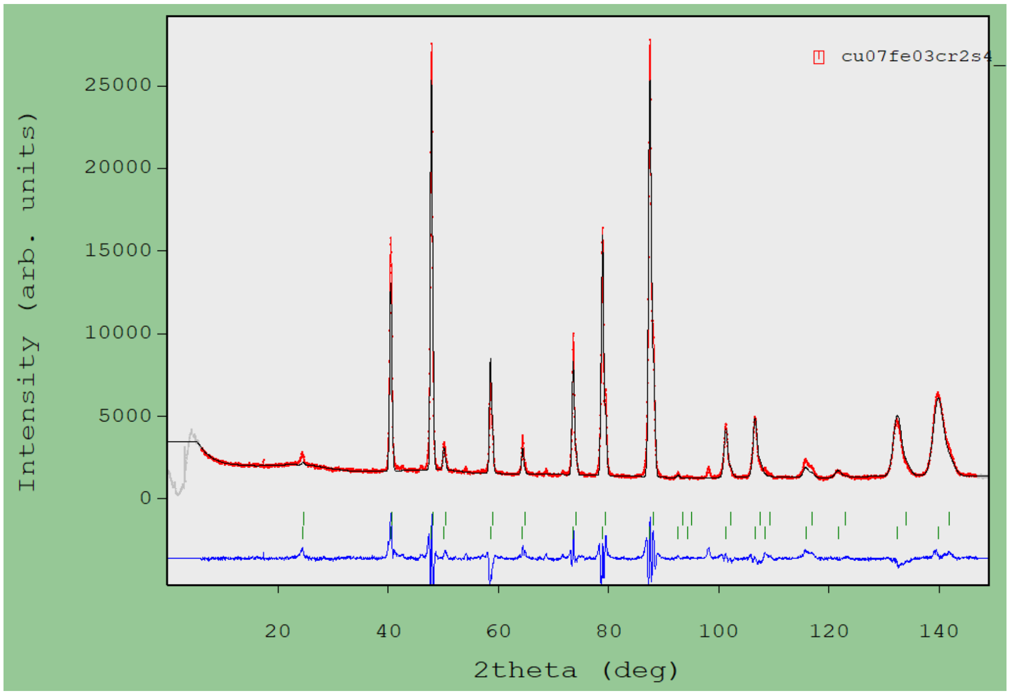

Figure 2. Rough refinement of the powder neutron diffraction pattern obtained for $Fe_{0.3}Cu_{0.7}Cr_2S_4$ to estimate phase composition. As visible by the peak splitting at large scattering angles, the data is best fitted with the combination of 30% of the pure phase $FeCr_2S_4$ and 70% of the pure phase $CuCr_2S_4$.

Conclusion

Due to unexpected problems with sample preparation, only mixtures of the pure compounds $FeCr_2S_4$ and $CuCr_2S_4$ were measured and the experiment did not yield new information. Based on the earlier preparation of homogenous polycrystalline samples by our team [2], phase separation was not anticipated and was not noticed by laboratory characterization.

References

[1] E. Sadrollahi, *$Fe_{1-x}Cu_xCr_2S_4$ Studied by Mössbauer Spectroscopy and Muon Spin Rotation and Relaxation*, PhD Thesis, TU Braunschweig (2018) DOI: 10.24355/dbbs.084-201811081221-0

[2] V. Tsurkan et al., J. Phys. Chem. Solids 66, 2040 (2005)