Experimental report

15/04/2021

Proposal: **5-32-895** **Council:** 10/2019

Title: Short-range correlations in the S = 1/2 square-lattice antiferromagnets Sr2CuTeO6 and Sr2CuWO6

Research area: Physics

This proposal is a continuation of 5-31-2638

Main proposer: **Ellen FOGH**

Experimental team: Lucile MANGIN-THRO
Ellen FOGH
Otto MUSTONEN
Helen WALKER

Local contacts: Lucile MANGIN-THRO

Samples: Sr2CuWO6
Sr2CuTeO6

Instrument	Requested days	Allocated days	From	To
D7	5	4	31/08/2020	04/09/2020

Abstract:

We propose a 5 day experiment at D7 to study the spin correlations in the short-range correlated states above TN in the compounds Sr2CuTeO6 and Sr2CuWO6. These are square lattice systems with J1 along the side of the square and J2 along the diagonal. The Te compound has a dominant J1 interaction and orders in a Néel state whereas the W compound has a dominant J2 interaction accompanied by columnar antiferromagnetic order. Thus, long-range order occurs in these compounds below TN = 29K and TN = 24K but above the ordering temperature significant 2D magnetic correlations persist as seen in a broad maximum in the susceptibility and with inelastic neutron scattering. A spin-liquid-like state has previously been observed in the Sr2CuTe(1-x)WxO6 solid solution series. However, a previous D7 experiment on Sr2CuTe(1-x)WxO6 with x = 0.2, 0.5, showed 2D correlations that might correspond to the correlations seen in the parent compounds above the ordering temperature. With the proposed experiment we aim to unravel if the low-temperature states for x = 0.2, 0.5 are simply an extension of the short-range correlated states in the parent compounds or whether they are of a different nature.

Short-range correlations in the S = 1/2 square-lattice antiferromagnets Sr_2CuTeO_6 and Sr_2CuWO_6

Proposal no: 5-32-895
Beamtime: D7: 4 days (31-08-2020 to 04-09-2020)
Experiment team: Otto Mustonen (University of Sheffield), Helen Walker (ISIS neutron and muon source), Ellen Fogh (École Polytechnique Fédérale de Lausanne)
Local contacts: Lucile Mangin-Thro
Comments: The experiment was carried out remotely due to Covid travel restrictions

The S = 1/2 Heisenberg frustrated square-lattice model (FSL) is one of the simplest models in frustrated magnetism. In this model magnetic frustration arises from the competition of antiferromagnetic nearest-neighbor J_1 (side of the square) and next-nearest neighbor J_2 (diagonal) interactions. Anderson [1] predicted that FSL systems could host a quantum spin liquid (QSL) ground state, when magnetic frustration is maximized at J_2/J_1 = 0.5. However, no known compound exists in this regime of the phase diagram and a square-lattice QSL has never been observed.

Sr_2CuTeO_6 and Sr_2CuWO_6 are realizations of the FSL model with Cu^{2+} (S = 1/2) cations forming the square lattice [2,3], see Figs. 1a)+b). Despite being isostructural, the magnetic interactions are completely different due to orbital hybridization effects: Sr_2CuTeO_6 has a dominant J_1 interaction (Néel magnetic structure), whereas in Sr_2CuWO_6 the J_2 interaction dominates (columnar magnetic structure). The QSL state is expected between these two extremes, and we have observed a spin-liquid-like ground state in the $Sr_2CuTe_{1-x}W_xO_6$ solid solution series between these two parent compounds, see Fig. 1c). In the composition range x = 0.2-0.6 magnetic order disappears completely down to 19mK, and specific heat suggest gapless excitations consistent with a QSL ground state [4,5].

In a previous D7 experiment (proposal no.5-31-2638) we studied two compositions x = 0.2 and x = 0.5 in the spin-liquid-like region of the $Sr_2CuTe_{1-x}W_xO_6$ phase diagram. The extracted magnetic diffuse scattering was fitted using SPINVERT [6], see Figs. 2a)+b). Despite both compositions having a dynamic magnetic ground state, the spin correlations in these two compounds are quite different. In x = 0.2, the nearest-neighbor spin correlations are strongly

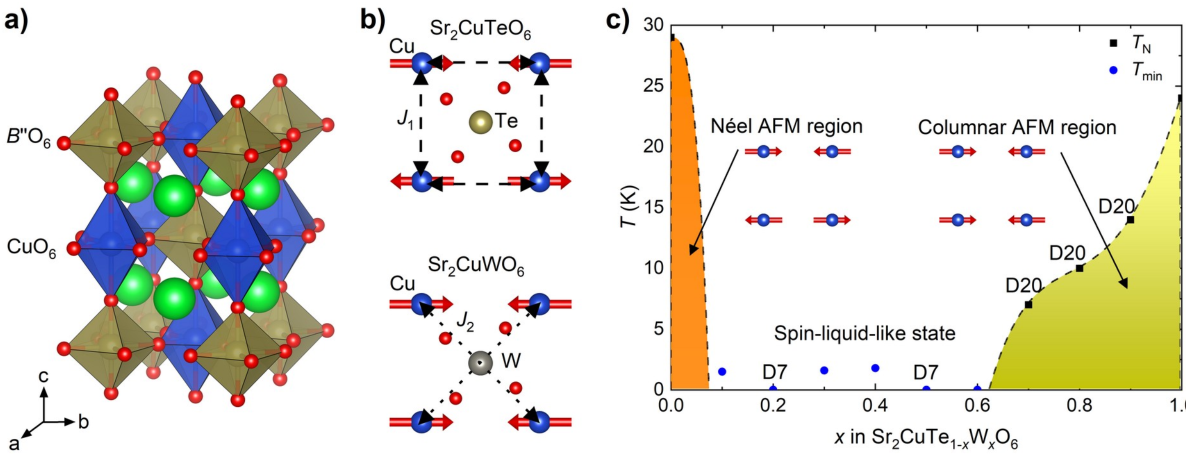

Figure 1: *a) Crystal structure of $Sr_2CuTe_{1-x}W_xO_6$. The Cu^{2+} square-lattice is formed in the ab plane. b) The Néel magnetic structure of Sr_2CuTeO_6 with dominant J_1, and the columnar magnetic structure of Sr_2CuWO_6 with dominant J_2. c) Phase diagram for $Sr_2CuTe_{1-x}W_xO_6$. For x = 0.2-0.6 magnetic order disappears and a spin-liquid-like state is observed. Figures are from Ref. 4.*

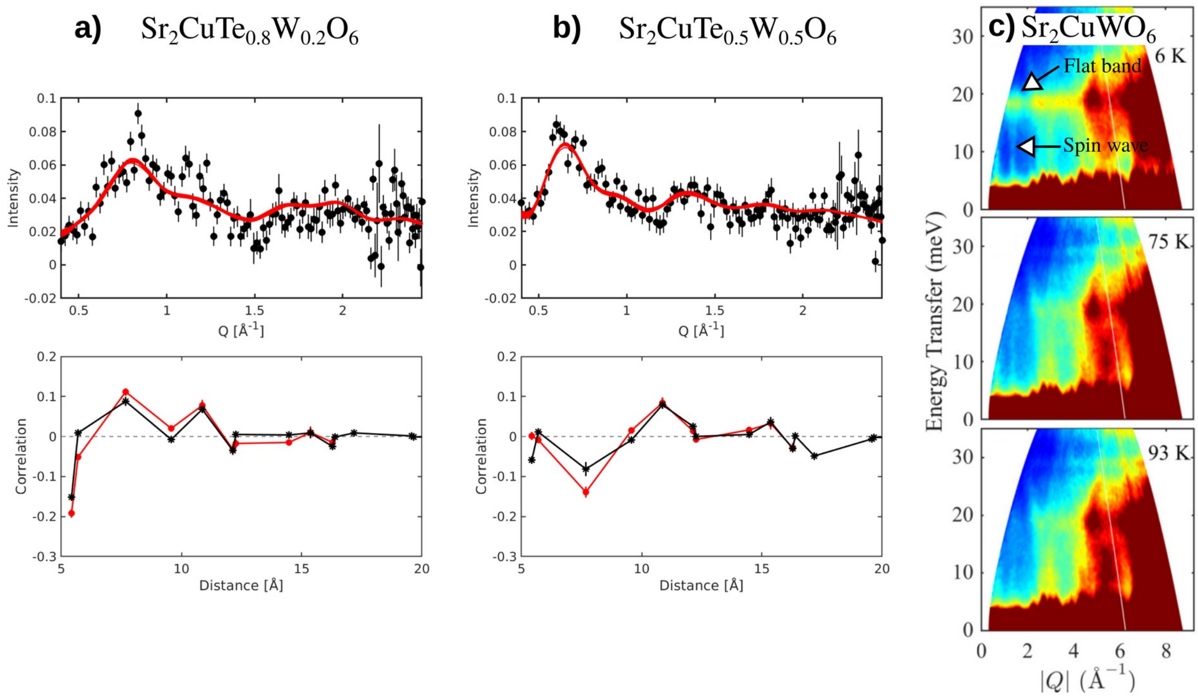

Figure 2: *a) and b) Measured diffuse magnetic scattering of x = 0.2 and x = 0.5 on D7 with preliminary analysis using SPINVERT. The bottom panels show the obtained spin correlations (red) in comparison with those resulting from Monte Carlo simulations (black). c) Inelastic measurements on* Sr_2CuWO_6 *as measured with MERLIN at ISIS, UK [3]. Evidence of spin-waves persist up to 75K, i.e well above* T_N *= 24K.*

antiferromagnetic, whereas in $x = 0.5$ the next-nearest neighbor spin correlation is strongly antiferromagnetic, and the nearest-neighbor one is weakly ferromagnetic. The spin correlations obtained from SPINVERT [6] qualitatively agree with spin correlations obtained from Monte Carlo simulations of a model Hamiltonian on the square lattice [7]. These results suggest that the dynamic ground state might not be a true QSL, but an extension of dynamic 2D correlations observed in the parent phases above their Néel temperatures.

Long-range magnetic order in the parent compounds Sr_2CuTeO_6 and Sr_2CuWO_6 occurs at T_N = 29K and T_N = 24K, respectively. This is caused by weak out-of-plane interactions. Above the Néel temperature significant 2D magnetic correlations persist as evidenced by a broad maximum in magnetic susceptibility (with $T_{max} >> T_N$). Further evidence for a short-range correlated state is provided by inelastic neutron scattering experiments, which reveal magnetic excitations up to at least 60K in Sr_2CuTeO_6 and 75K in Sr_2CuWO_6 [3], see Fig. 2c).

We speculated, based on the spin correlations, that the dynamic magnetism observed in x = 0.2 might be closely related to the 2D correlated state in Sr_2CuTeO_6, and that of x = 0.5 to the 2D correlated state of Sr_2CuWO_6. We therefore set out to investigate the short-range correlations in the two parent compounds by performing a diffuse diffraction experiment on D7 above the Néel temperatures. Powder samples of around 19g were used and dataset at base temperature as well as at 40, 60 and 100K were collected for the x = 0 compound and for 40K for the x = 1 compound.

Figure 3a) shows all the data collected for the x = 0 compound. At 1.5K there are clear Bragg peaks corresponding to Néel antiferromagnetic order. At 40K, these peaks have disappeared but a broad bump around the first peak position at 0.8$Å^{-1}$ is still evident. At yet higher temperatures this bump gradually broadens and is finally no longer there at 100K. This agrees well with the previous INS measurements [3], although on the x = 1 compound. However, the two compounds have similar size exchange parameters and T_N and are thus expected to have a similar temperature-dependent behavior.

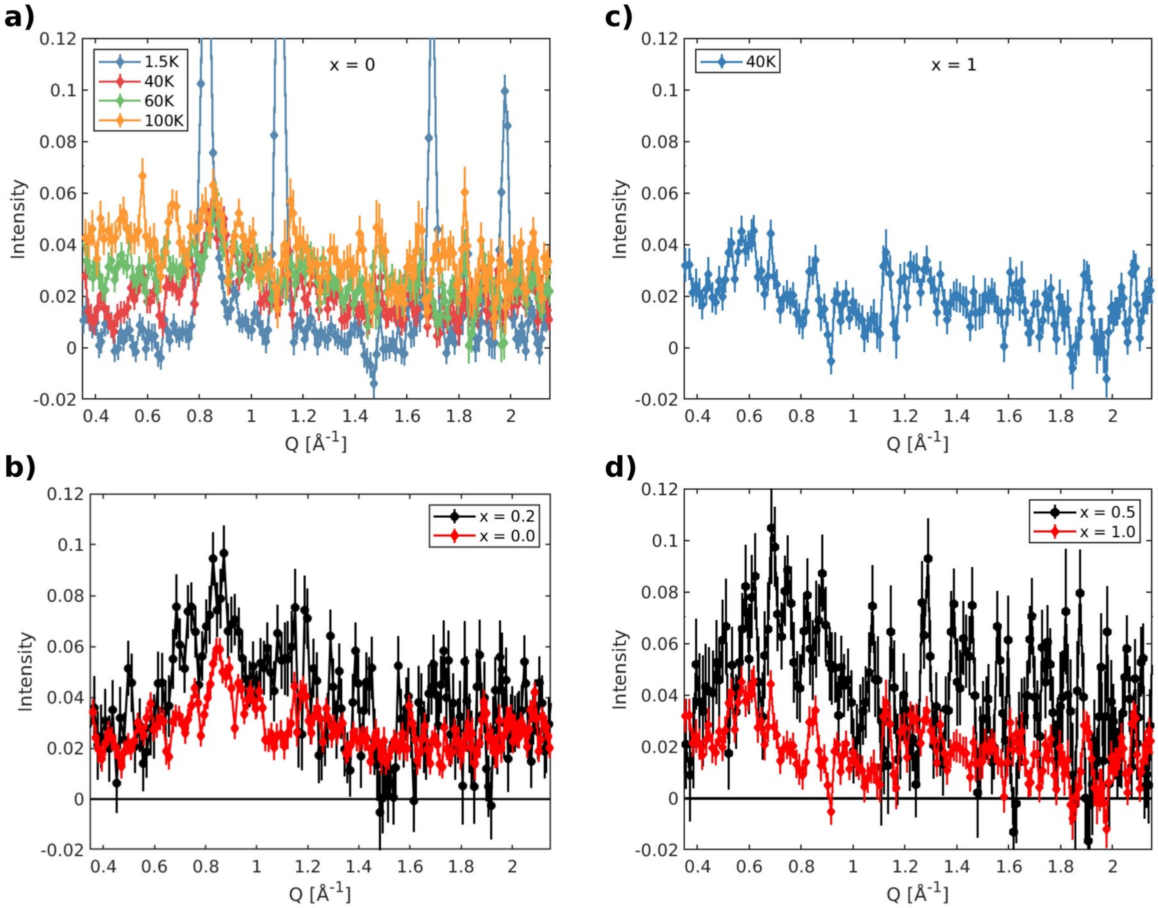

Figure 3: a) Comparison of all dataset collected for the x = 0 compound. b) Diffuse scattering for the x = 0 compound at 40K compared with that for the x = 0.2 compound at 1.5K. c) and d) Data for the x = 1 compound at 40K by itself and compared with the x = 0.5 compound at 1.5K, respectively.

Our previous D7 measurements for $x = 0.2$ at 1.5K are compared with those for $x = 0$ at 40K in Fig. 3b). The two dataset show similar trends. This means that the short-range correlations observed in the $x = 0.2$ compound are indeed similar to those above T_N for $x = 0$. Results for the $x = 1$ compound at 40K are shown in Fig. 3c) and compared with our previous measurements on the $x = 0.5$ compound at 1.5K in Fig. 3d). For $x = 1$ at 40K, the weight of the scattering intensity is around 0.6Å^{-1}, corresponding to the columnar antiferromagnetic order. For $x = 0.5$ at 1.5K the weight has also shifted towards 0.6Å^{-1} as compared to for $x = 0.2$ where the highest intensity is found around 0.8Å^{-1}. However, for $x = 0.5$ there is still significant intensity at 0.8Å^{-1}. Thus the short-range correlations found in the $x = 0.5$ compound are most likely a mixture of those found above T_N in both parent compounds.

A more thorough analysis of the correlations is ongoing using SPINVERT [6].

[1] P. W. Anderson, *Science* **235**, 1196 (1987).
[2] P. Babkevich et al., *Phys. Rev. Lett.* **117**, 237203 (2016).
[3] H. C. Walker et al., *Phys. Rev. B* **94**, 064411 (2016).
[4] O. Mustonen et al., *Nat. Commun.* **9**, 1085 (2018).
[5] O. Mustonen et al., *Phys. Rev. B* **98**, 064411 (2018).
[6] J. A. M. Paddison et al., *J. Phys. Condens. Matter* **25**, 454220 (2013).
[7] V. M. Katukuri et al., *Phys. Rev. Lett.* **124**, 077202 (2020)