Experimental report

13/09/2019

Proposal: 5-31-2638 **Council:** 10/2018

Title: Bond disorder on a two-dimensionalsquare lattice of the antiferromagnetic Sr2Cu(W,Te)O6

Research area: Physics

This proposal is a new proposal

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Samples: Sr2Cu(W,Te)O6

Instrument	Requested days	Allocated days	From	To
D20	5	2	14/06/2019	16/06/2019
D7	5	5	30/07/2019	04/08/2019

Abstract:

In this proposal we wish to examine the nature of the magnetic order in Sr2Cu(W,Te)O6. This compound has attracted a lot of interest lately in that by substituting W for Te one can tune between Neel to columnar antiferromagnetic (AFM) order. The end compounds containing only W or Te have been investigated in detail but no neutron diffraction data exists for the intermediate dopings. Our aim is to to combine D20 and D7 measurements to elucidate the nature of ground state and the microscopic interactions as Te -> W for which we have developed detailed theoretical models.

Bond disorder on a two-dimensional square lattice of the antiferromagnetic $Sr_2Cu(W,Te)O_6$

Proposal no: 5-31-2638
Beamtime: D20: 2 days, D7: 5 days
Experiment team: Otto Mustonen (University of Sheffield), Helen Walker (ISIS neutron and muon source), Ellen Fogh (École Polytechnique Fédérale de Lausanne)
Local contacts: Thomas Hansen, Lucile Mangin-Thro

The S = 1/2 Heisenberg frustrated square-lattice model is one of the simplest models in frustrated magnetism. Anderson [1] predicted that these systems could host a quantum spin liquid ground state (QSL), but it has never been previously observed. We have performed a neutron scattering study of such square-lattice QSL candidate system $Sr_2CuTe_{1-x}W_xO_6$. The phase diagram of the system, based on muon spin rotation and relaxation measurements, is presented in Fig. 1 [2]. Néel magnetic order is observed in Sr_2CuTeO_6 (x = 0) below T_N = 29 K. When adding tungsten, magnetic order is suppressed for a wide region of x = 0.2-0.6, and a spin-liquid-like state is observed. Magnetic order is observed again for x = 0.7-1, where Sr_2CuWO_6 (x = 1) is known to have columnar antiferromagnetic order below T_N = 24K.

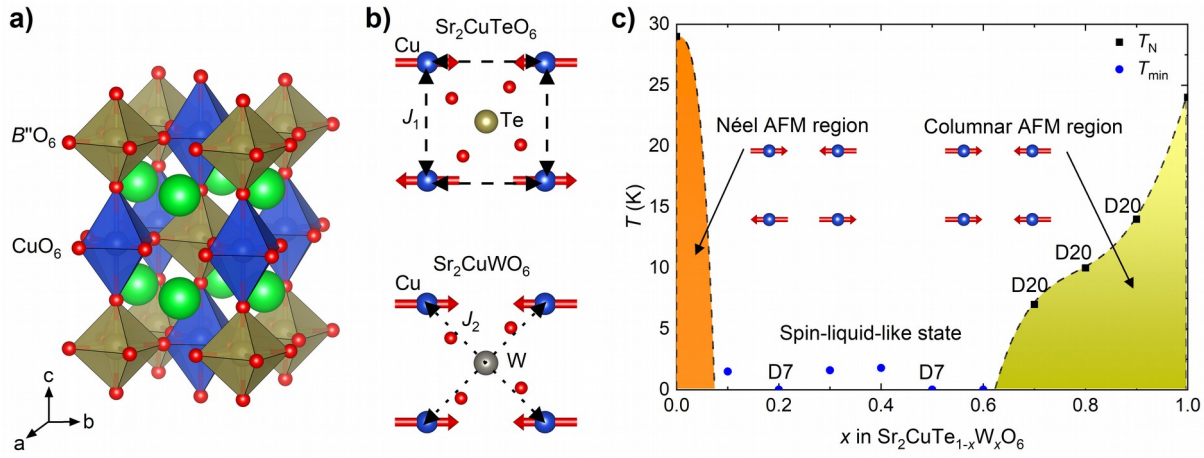

Figure 1: *a) Crystal structure of $Sr_2CuTe_{1-x}W_xO_6$. The Cu^{2+} square-lattice is formed in the ab plane. b) The Néel magnetic structure of Sr_2CuTeO_6 with dominant J_1, and the columnar magnetic structure of Sr_2CuWO_6 with dominant J_2. c) Phase diagram for $Sr_2CuTe_{1-x}W_xO_6$. For x = 0.2-0.6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

High-flux neutron diffraction experiments were performed on D20 on samples with x = 0.7, 0.8 and 0.9, see Fig. 2. While previous muon spin rotation experiments established the presence of magnetic order in these samples, our aim was to establish the type of magnetic order. The magnetism in these compounds is very weak, but magnetic Bragg peaks could be observed when 30K nuclear data was subtracted from the low-temperature 2K nuclear+magnetic data. In x = 0.9, a number of clear reflections corresponding to the propagation vector k = [0 1/2 1/2] were observed. This indicates that the in-plane magnetic structure is columnar (first two Miller indices), whereas the coupling between planes along c (last Miller index) is antiferromagnetic. This is identical to magnetic structure of Sr_2CuWO_6 (x = 1). The refined Cu^{2+} moment for x = 0.9 was 0.45μ_B, which is weaker than the 0.6μ_B observed for Sr_2CuWO_6.

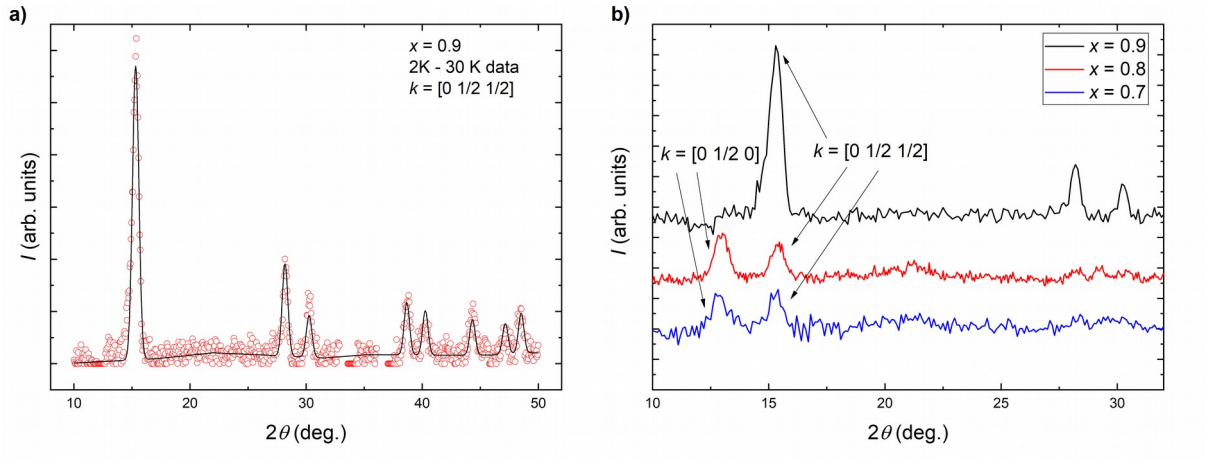

Figure 2: *a) Magnetic neutron scattering data for x = 0.9 at 2K (30K data subtracted). The magnetic structure can be refined with the propagation vector k = [0 1/2 1/2] with a Cu^{2+} moment of 0.46μ_B along a. b) Magnetic neutron scattering data for x = 0.9, 0.8 and 0.7. In the latter two an additional strong reflection corresponding to k = [0 1/2 0] is observed in addition to k = [0 1/2 0].*

The magnetic peaks in x = 0.8 and x = 0.7 are much weaker than in x = 0.9. Surprisingly, a peak corresponding to another propagation vector, k = [0 1/2 0], is observed in these compounds. This propagation vector also corresponds to columnar magnetic order in the square plane, but now with ferromagnetic stacking of layers along c. The presence of reflections corresponding to both propagation vectors suggests, that Te^{6+} doping disrupts the interactions along c in this compounds, which are ultimately responsible for 3D magnetic order.

To complement these measurements, we performed polarized neutron scattering experiments on D7 for samples with composition x = 0.2 and 0.5. These are known to have a spin-liquid-like ground state. From calculations (Fig. 3a) we expected a shift in scattering intensity from the |Q| ~ 0.6Å^{-1} to |Q| ~ 0.8Å^{-1} corresponding to columnar and Néel order respectively and this is indeed what we observe in the diffuse scattering signal (Figs. 3b+c). Using SPINVERT [3] to fit the obtained magnetic diffuse scattering signal yields correlations as a function of bond length as seen in the lower part of Figs. 3b+c. For x = 0.2, the nearest-neighbor correlation is strongly antiferromagnetic, similar to the Néel state. For x = 0.5, the nearest-neighbor interaction is weak but the next-nearest neighbor correlation is antiferromagnetic,

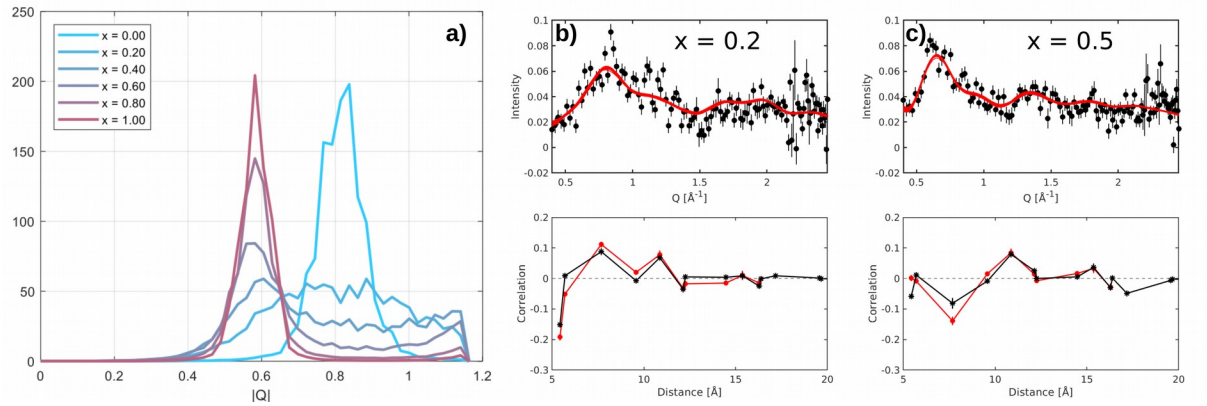

Figure 3: *a) Calculated powder-averaged magnetic intensities in the (H,K) scattering plane. Measured diffuse magnetic scattering of x = 0.2 and x = 0.5 on D7 with preliminary analysis using SPINVERT. The spin correlations (red) of these compounds are distinctly different, but qualitatively similar to Monte Carlo simulations (black).*

corresponding to the columnar phase. We compare these with Monte Carlo simulations performed using a model Hamiltonian [4] and a reasonable agreement has been obtained.

[1] P. W. Anderson, Science 235, 1196 (1987).

[2] O. Mustonen et al., Nat. Commun. 9, 1085 (2018).

[3] J. A. M. Paddison et al., J. Phys. Condens. Matter 25, 454220 (2013).

[4] V. M. Katukuri et al., arxiv:1902.09376.