Experimental report

15/07/2021

Proposal: **4-01-1655** **Council:** 10/2019

Title: Magnetic excitation spectrum at the deconfined quantum critical pointof the Shastry-Sutherland compound, SrCu2(BO3)2

Research area: Physics

This proposal is a new proposal

Main proposer: **Ellen FOGH**

Experimental team: Mohamed ZAYED
Irina SAFIULINA
Martin BOEHM
Ursula Bengaard HANSEN
Alexandre IVANOV
Jian Rui SOH
Ellen FOGH
Gaetan GIRIAT
Stefan KLOTZ
Louis AMAND

Local contacts: Martin BOEHM
Alexandre IVANOV
Andrea PIOVANO

Samples: SrCu2(BO3)2

Instrument	Requested days	Allocated days	From	To
IN8	7	6	01/06/2021	07/06/2021
THALES	3	3	08/06/2021	10/06/2021
IN3	1	3	18/05/2021 31/05/2021	20/05/2021 02/06/2021

Abstract:

We propose a 7 day inelastic neutron scattering experiment at IN8 to verify the predicted deconfined quantum critical point in the Shastry-Sutherland compound, SrCu2(BO3)2. If the experiment is succesful, this would be the first ever physical realization of such a critical point. The Shastry-Sutherland lattice consists of spin pairs (dimers) embedded in a square lattice with inter-dimer coupling, J, and intra-dimer coupling, J'. It has an exact dimer product ground state for J'/J <= 0.675. Upon increasing the ratio, J'/J, the system enter a plaquette singlet phase and finally the Néel state. SrCu2(BO3)2 is unique since it is topologically equivalent to the Shastry-Sutherland lattice and with the possibility to tune J'/J by applying pressure. Therefore, SCBO presents an amazing experimental playground for theoretical predictions. One such prediction is the existence of a so-called deconfined quantum critical point at the phase transition between the plaquette phase and Néel state at 4.0GPa. This critical point has a distinct footprint in the magnetic excitation spectrum and it is exactly what we aim to investigate with the proposed experiment.

Magnetic excitation spectrum at the deconfined quantum critical point of the Shastry-Sutherland compound, $SrCu_2(BO_3)_2$

Proposal no: 4-01-1655
Beamtime: IN8 (8 days) + ThALES (3 days)
Experiment team: Mohamed Zayed (Carnegie Mellon University Qatar), Gaétan Giriat and Ellen Fogh (École Polytechnique Fédérale de Lausanne)
Local contacts: Andrea Piovano, Martin Boehm

The Shastry-Sutherland (SS) lattice consists of spin pairs (dimers) embedded in a square lattice (see Fig. 1a) and with inter-dimer coupling, J, and intra-dimer coupling J’. It has an exact dimer product ground state for J’/J ≤ 0.675 [1]. Upon increasing the ratio of J’/J, the system goes through a quantum phase transition to a plaquette singlet state followed by a transition to a Néel phase [2]. $SrCu_2(BO_3)_2$ (SCBO) is a unique material since it is topologically equivalent to the SS lattice [3]. With J’/J ~ 0.6 close to the critical point, SCBO presents remarkable experimental testing grounds for the SS model. The ratio J’/J may be tuned by applying pressure and the resulting phase diagram resembles that theoretically predicted for the SS model [4] (see Fig 1b).

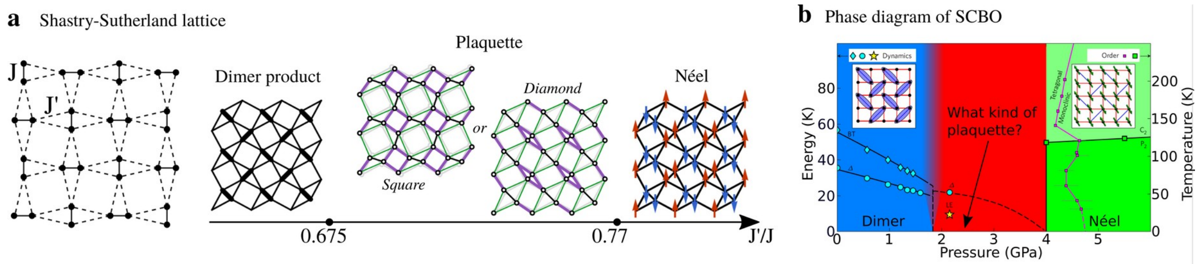

Figure 1: *(a) Illustration of the SS lattice with inter-dimer and intra-dimer coupling, J and J’, respectively, as well as the theoretically predicted phase diagram [5]. (b) Experimental phase diagram of SCBO under pressure [4].*

In this experiment we focused on magnetic excitations in the plaquette and Néel phases with transitions at 1.8GPa and 4.0GPa, respectively. It was performed with a 60mg single crystal of SCBO in a Paris-Edinburgh pressure cell loaded first to 3.5GPa (plaquette phase) and then increased to 5.5GPa (Néel phase). The crystal was oriented with (HK0) in the horizontal scattering plane.

In the plaquette phase there is evidence in the specific heat of a transition around 2K [6] so we cooled down by filling liquid He straight into the cryostat. This way we were able to maintain a temperature of 1.5K for roughly 12h and with the sample above the He level for 7 of those. The inelastic neutron data we collected at 1.5K and 4.5K is shown in Fig. 2 and it is clear that cooling down below 2K is not necessary in order to see the excitations in the plaquette phase. We performed a number of energy scans at fixed Q and observed excitations at low energies close to the elastic line but also increased intensity for the entire spectrum. Unfortunately, the He gas compressor maintaining the pressure in the cell failed and when eventually recovering the pressure we decided to go higher and investigate the Néel phase. Therefore, our results from the plaquette phase are by far complete so we will apply for more beamtime in the future.

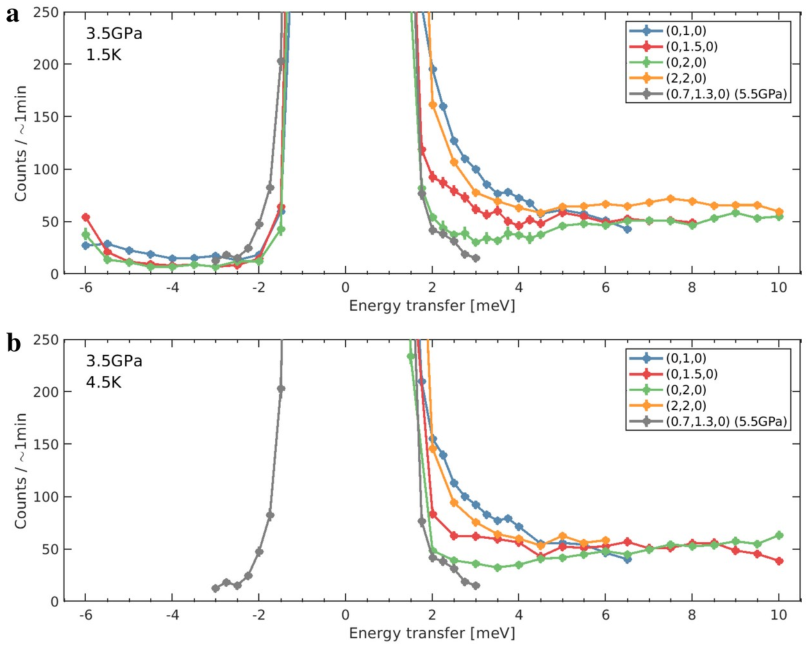

Figure 2: *Energy scans at various positions in reciprocal space in the plaquette phase at (a) 1.5K and (b) 4.5K. Clearly, there is nothing gained by cooling down to 1.5K.*

Figure 3 shows our results from IN8 in the Néel phase. As expected we see spinwaves from an antiferromagnet. Some analysis work is still to be done in order to understand how the behave. Energy gaps from ThALES, see Fig. 4, show that the system has a gap of around 1.9meV and this is unexpected from the isotropic SS model. However, SCBO does deviate from the SS model. For example, the planes made up by the Cu^{2+} ions are buckled and not flat which allows for the Dzyaloshinskii-Moriya interaction. Moreover, above 4.5GPa SCBO goes from tetragonal crystal ctructure to monoclinic [7] and the SS model is possibly no longer a good description for the physical system.

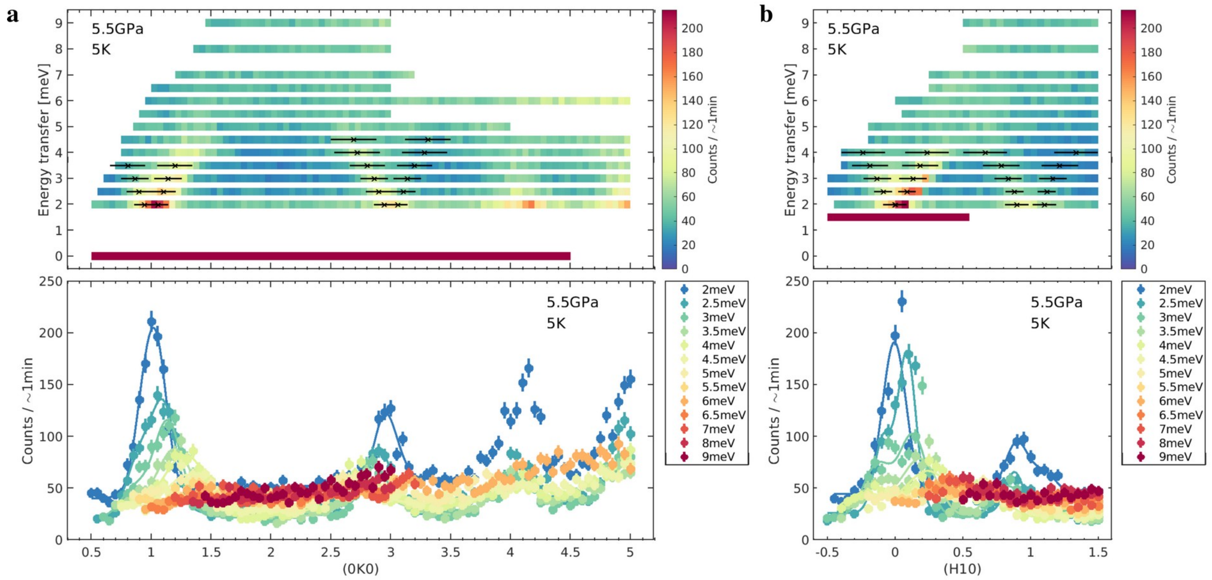

Figure 3: *Constant energy scans along (0K0) and (H10) as measured at IN8 in the Néel phase. The solid lines in the bottom panels are fits to two Gaussians positioned with equal distance either side of (010), (030) and (110). The fitted positions are plotted with the black crosses in the colorplots in the top panels.*

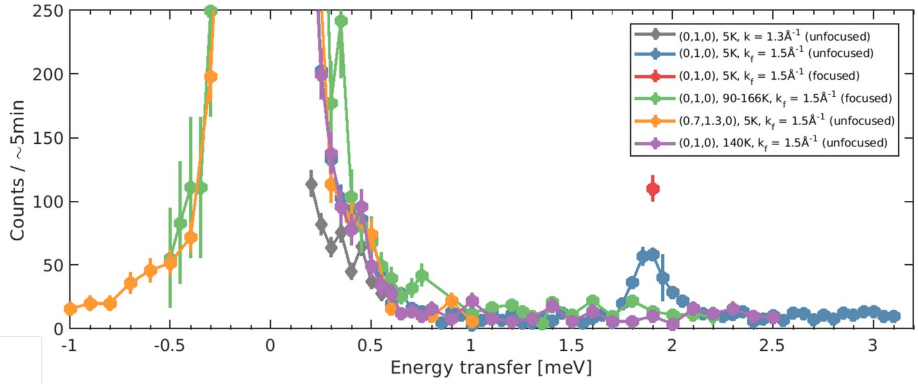

Figure 4: *Energy scan at (010) at various temperatures as measured at ThALES in the Néel phase. The spinwave is gapped around 1.9meV.*

In conclusion, our experiment showed that doing inelastic neutron scattering experiments on 60mg sample of a S = ½ system inside a Paris-Edinburgh cell is indeed feasible. There is obviously still a lot of analysis and interpretation to do with the measured data both in the plaquette phase and in the Néel phase. We also need to investigate the plaquette phase more, ideally at an instrument with better energy resolution than IN8 in order to look at the low-energy excitations. We are also lacking information about the magnetic structure in the Néel phase as this study was focused on the excitations. However, performing another experiment to collect magnetic Bragg peak intensities would be a valuable supplement to the current dataset.

[1] B. S. Shastry and B. Sutherland, Physica **108B**, 1069-1070 (1981)
[2] A. Koga and N. Kawakami, Phys. Rev. Lett. **84**, 4461–4464 (2000)
[3] S. Miyahara and K. Ueda, Phys. Rev. Lett. **82**, 3701 (1999)
[4] M. E. Zayed et al. Nature Physics **13**, 962 EP (2017)
[5] J. Y. Lee et al., *Phys. Rev. B* **9**, 041037 (2019)
[6] J. Guo et al., *Phys. Rev. Lett.* **124**, 206602 (2020)
[7] M. E. Zayed et al., *Solid State Comm.* **186**, 13-17 (2014)