In this study we have investigated the low-dimensional correlated spin system SrCu$_2$(BO$_3$)$_2$ using ambient-pressure muon spin rotation/relaxation ($\mu^+\text{SR}$). The zero-field data are similar to previously published data, but in addition, they give an even clearer sign of the two low-temperature transitions ($T_1 \approx 3$ K and $T_2 \approx 7$ K), which is fully consistent with inelastic neutron scattering (INS) measurements. Longitudinal field (LF) data clearly show that the copper spins are highly dynamic and a saturation of the low-temperature relaxation rate indicate that these are indeed two-dimensional (2D) quantum spin fluctuations.

KEYWORDS: Low-dimensional spin system, Shastry-Sutherland model, Dzyaloshinskii-Moriya interaction

1. Introduction

The holy grail of condensed matter physics is clearly the understanding and realization of room-temperature superconductivity. Even though the overall understanding of this exotic phenomena has improved slowly over the years, we still have a long way to go. One of the key discoveries in this field was clearly the presence of ($S = 1/2$) spin-correlations within the copper oxide planes of the cuprate high-temperature superconductors (HTSC) [1]. Following such results both theoretical and experimental physicists has invested a huge efforts in the general understanding of low-dimensional correlated electron systems [2–10]. To continue this development also solid state chemists have discovered and synthesized a wide range of new compounds, especially materials that contains square CuO$_4$ planes. Among the more intriguing and well studied ones we find the title compounds i.e. SrCu$_2$(BO$_3$)$_2$ (SCBO, from hereon). SCBO has a tetragonal unit cell with lattice parameters $a = b = 8.995$ Å and $c = 6.649$ Å at room temperature. It consists of alternately stacked Sr- and CuBO$_3$-planes. Neighboring pairs of planar rectangular CuO$_4$ forms spin dimmers, which are connected orthogonally by a triangular BO$_3$ (see also Fig. 1).

The Shastry-Sutherland model [11] has an “exact” dimer ground state (a direct product of spin singlets on the nearest neighbor bonds), which comes as a result from the atypical exchange geometry. For SCBO this is realized by the fact that the Cu$^{2+}$ spins ($S = 1/2$) are arrange in a 2D lattice...
with strongly coupled dimers ($J = 7.3$ meV) with a geometrically frustrated inter-dimer coupling ($J' = 4.7$ meV). This result in almost completely isolated dimers and a triplet gap of $E = 3$ meV. Further, inelastic neutron scattering (INS) studies have revealed anisotropic Dzyaloshinskii-Moriya interactions and a tiny interlayer coupling $J'' = 0.7$ meV [12]. We have indeed already performed experimental investigation of the model material using inelastic neutron scattering (INS) and have shown that it is possible to tune SCBO by application of hydrostatic pressure out of the exact dimer phase to a suggested plaquette singlet state and eventually to an antiferromagnetic ordered phase. These transitions occur at $p_{c1} \approx 18$ kbar and $p_{c2} \approx 40$ kbar, respectively [13–16].

![Crystallographic data and atomic structure of SrCu$_2$(BO$_3$)$_2$ showing the flat copper oxide (ab-)planes as well as emphasized Cu-Cu dimers.](image)

**Fig. 1.** Crystallographic data and atomic structure of SrCu$_2$(BO$_3$)$_2$ showing the flat copper oxide (ab-)planes as well as emphasized Cu-Cu dimers.

### 2. Experimental Details

Large (several centimeters long rods) high-quality single crystalline samples of SCBO were grown by the traveling solvent floating zone technique following a similar receipt and methods presented in reference [17]. Part of such a single crystalline rod was cut off and ground into a fine powder. The reason that we used this method to obtain a pure powder sample is that "as prepared" powder samples used for previous studies have shown to contain a small fraction of impurity phases [13].

For the $\mu^+\text{SR}$ experiment the ground powder sample was placed into a small envelope made of $50 \mu$m thin Al-coated Mylar tape and then attached to a low-background fork-type sample holder. In order to make certain that the muons stopped primarily inside the sample, we ensured that the side facing the muon beamline was covered only by a single layer of Mylar tape. Subsequently, $\mu^+\text{SR}$ spectra were recorded at the Swiss Muon Source ($\text{S}\mu\text{S}$), Paul Scherrer Institut, Villigen, Switzerland. By using the surface muon beamline $\pi\text{M3}$ and the GPS spectrometer, zero-field ($ZF$) and weak transverse-field ($w\text{TF}$) data were collected for $1.5 \text{ K} \leq T \leq 50 \text{ K}$. The experimental setup and techniques were described in detail elsewhere [18].
3. Results & Discussion

μSR time spectra recorded in ZF at $T = 1.6$ K show no clear oscillations but rather a slow exponential decay indicating that SCBO enters into either a static spin-glass state (resulting in a wide field-distribution) or that the electron spins are dynamic even at this low temperature. The ZF data was found to be well fitted to a slow decaying stretched-exponential function:

$$A_0 P_{ZF}(t) = A \cdot e^{-(\lambda t)^\beta},$$

(1)

Here $A$ is the asymmetry, $\lambda$ the relaxation rate and $\beta$ the so-called critical exponent. ZF spectra were acquired as a function of temperature $1.5$ K $\leq T \leq 50$ K. Data in the entire temperature range was well fitted to Eq. 1 and the fitting results are shown in Fig. 2. For both $\beta(T)$ and $\lambda(T)$ the present data reveals two distinct transitions (or crossover temperatures) at $T_1 \approx 3$ K and $T_2 \approx 7$ K. Such transition temperatures agrees well with previously published INS data [14,17]. More surprisingly is that in previously published $\mu^+\text{SR}$ data on SCBO [19] only the $T_1$ transition was present while $T_2$ was absent. This could be an indication of difference in sample quality or related to the use of slightly different fitting functions.

For the present data it is also clear that the critical exponent decreases from $\beta = 2$ (i.e. Gaussian relaxation) at high temperatures, to $\beta = 0.67$ for low temperatures. Here $\beta$ is known to depend on the effective dimensionality ($d$) of the system above (but in the vicinity of) a transition according to $\beta = \frac{d_c}{d+1}$. Here, that $\beta \rightarrow 0.67$ for clearly indicates the drive towards a 2D spin system in SCBO at low temperatures, as could be expected.

To distinguish between the spin-glass state or the dynamic spin scenario for SCBO we have also performed LF scans at lowest temperature ($T = 1.6$ K). The raw data is presented in Fig. 3(a) and the fitting results (Eq. 1) $\lambda(T)$ are shown in Fig. 3(b). First of all it is clear that small applied fields ($\text{LF} = 5-300$ G) does not decouple the muon spin significantly. This is a clear indication that the relaxation is related to dynamic spins. This also shed further light on the fact that the relaxation rate ($\lambda$) increases with decreasing temperature and then saturates below $T_1$. This behaviour most likely indicates the presence of quantum spin fluctuations at low temperatures rather than related to thermal fluctuations, as suggested in previous $\mu^+\text{SR}$ studies [20]. Finally, it is also clear that a critical field around $\text{LF}_c \approx 750$ G strongly starts to decouple the spectra. However, even at highest measurable field $\text{LF} = 4$ kG, there is still a clear small persistent relaxation [see Fig. 3(b)]. In order to further understand the magnetic nature, particularly below $T_1$, we plan to measure the LF-spectra at several
Fig. 3. (a) Longitudinal field (LF) $\mu^+\text{SR}$ time spectra as a function of applied field. Solid lines are fits to Eq. 1. (b) Fitting results showing the temperature dependent relaxation rate $\lambda(T)$. Solid line is only a guide to the eye and the vertical dashed line indicate the critical field $LF_c \approx 750$ G where a rapid decoupling of the spectra mainly occurs.

temperatures to estimate the fluctuation rate $\nu$ and muon hyperfine parameter $\delta$ using the Redfield theory [21,22].

4. Summary

In summary we have used ambient-pressure muon spin rotation/relaxation ($\mu^+\text{SR}$) to study the low-dimensional compound SrCu$_2$(BO$_3$)$_2$ (SCBO). Our results give a clear view on the low-temperature changes in copper spin dynamics and display two cross-over temperatures ($T_1 \approx 3$ K and $T_2 \approx 7$ K) that are clear indications for ambient pressure precursors to high-pressure data from inelastic neutron scattering (INS) [13–16] as well as unpublished bulk measurements [23]. Finally, longitudinal field (LF) data clearly show that the copper spins are highly dynamic even at lowest measured temperature most likely due to two-dimensional (2D) quantum spin fluctuations.

5. Acknowledgements

This work was performed using the GPS muon spectrometer at the Swiss Muon Source ($S\mu S$) of the Paul Scherrer Institut (PSI), Villigen, Switzerland and we are thankful to the staff for their support. This research was supported by a Marie Skłodowska Curie Action, International Career Grant through the European Union and Swedish Research Council (VR), Grant No. INCA-2014-6426, as well as VR neutron project grant (BIFROST, Dnr. 2016-06955) and the Wenner Gren Foundation. Further, we also acknowledge funding provided by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japan, KAKENHI Grant No. 23108003, and Japan Society for the Promotion Science (JSPS) KAKENHI Grant No. 26286084. All images involving crystal structure were made with the DIAMOND software and the $\mu^+\text{SR}$ data was fitted using musrfit [24].

References