

# Spin correlation and field induced staggered magnetization in the 2D orthogonal dimer spin system $\text{SrCu}_2(\text{BO}_3)_2$

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## Abstract

We report the results of nuclear magnetic resonance (NMR) experiments on  $^{11}\text{B}$  nuclei in  $\text{SrCu}_2(\text{BO}_3)_2$ . The NMR shift at a magnetic field of 7 T shows a staggered component, which is ascribed to the buckling of the  $\text{CuBO}_3$  layers giving rise to the staggered magnetization and the staggered hyperfine coupling. The temperature variation of the orientation dependence of the shift indicates that the staggered magnetization grows significantly below 20 K, where the uniform magnetization decreases exponentially due to formation of the dimer singlet ground state.

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$\text{SrCu}_2(\text{BO}_3)_2$  has a two-dimensional network of orthogonal dimers of  $\text{Cu}^{2+}$  with spin- $\frac{1}{2}$ , which is equivalent to the Shastry–Sutherland spin model [1]. It has the dimer singlet ground state [2] and, in high magnetic fields, shows plateaus in the magnetization curve at  $\frac{1}{8}$ ,  $\frac{1}{4}$  and  $\frac{1}{3}$  of the fully saturated moment [3,4]. These plateaus have been considered as signature for localization of triplets forming superlattices. Recent NMR experiments [5] indeed confirmed such a superlattice in the  $\frac{1}{8}$ -plateau phase with a large rhomboid unit cell containing 16 spins per layer.

Although these properties can be largely understood by an isotropic Heisenberg spin model on the Shastry–Sutherland lattice, small perturbations specific to this material not included in the Shastry–Sutherland model cause interesting phenomena. For example, the splitting of the triplet excitation energies observed by ESR [6] and neutron scattering [7] have been ascribed to the Dzyaloshinskiy–Moriya (DM) interaction between two spins on neighboring dimers [7]. Another feature is the non-coplanar structure of the  $\text{CuBO}_3$  layers. The upper

and lower parts of Fig. 1 schematically show the structure of the  $\text{CuBO}_3$  layer viewed along the  $c$ -axis and the  $(1\bar{1}0)$ -direction, respectively. The latter shows buckling of the  $\text{CuBO}_3$  layers. The buckling invalidates the inversion symmetry at the center of a dimer bond, giving rise to the DM interaction between two spins on the same dimer. The buckling also results in tilting of the principal axes of the  $g$ -tensor. For the  $\text{Cu}(1)$  site in Fig. 1, for example, one principal axis is along the  $[1\bar{1}0]$  direction, while the other two make an angle  $\phi$  with the  $[001]$  ( $c$  direction) and the  $[110]$  directions, respectively. If the  $\text{CuBO}_3$  layers were flat  $\phi$  should be zero. The  $g$ -tensors for other Cu sites are obtained by successive rotation by  $90^\circ$  around the  $c$ -axis, yielding alternation of the principal axes. This means that the uniform magnetic field induces staggered magnetization with four sublattices. The intradimer DM interaction mentioned above also contributes to the staggered magnetization.

We have performed  $^{11}\text{B}$  NMR measurements to detect the field-induced staggered magnetization. Fig. 2 shows the hyperfine magnetic shift at the B(1) sites as a function of the direction of the external field rotated in the  $(1\bar{1}0)$  plane. By symmetry, the shift at the B(2) sites is obtained by interchanging  $\theta$  with  $-\theta$ . At 100 K, the extremal of the angular pattern is located  $9^\circ$  away from

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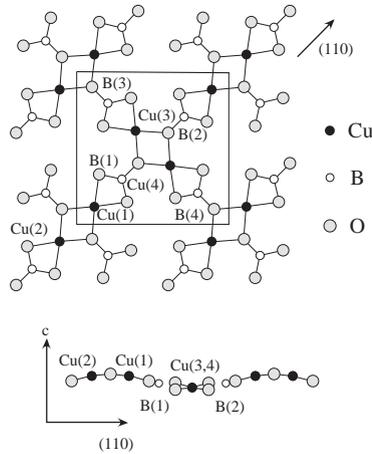


Fig. 1. Schematic structure of CuBO<sub>3</sub> layer in SrCu<sub>2</sub>(BO<sub>3</sub>)<sub>2</sub> viewed along the *c*-axis (upper panel). Atomic arrangement of CuBO<sub>3</sub> layer along the [1 1 0]-direction (lower panel, not in correct scale).

$\theta = 0$  or  $90$ . This deviation means different shift values for the B(1) and B(2) sites, i.e., a staggered component of the shift. Since the shift tensor is a product of the local magnetization and the hyperfine coupling tensor, staggered component in either or both of them will result in staggeredness of the shift. What is remarkable is that the position of the extremal moves rapidly below 20 K. Since the hyperfine coupling tensor should be nearly independent of temperature, such strong temperature dependence can be explained only by rapid growth of the staggered magnetization below 20 K. It is quite surprising that large staggered magnetization remains even at 2.5 K, where the uniform magnetization is negligibly small due to formation of the singlet ground state.

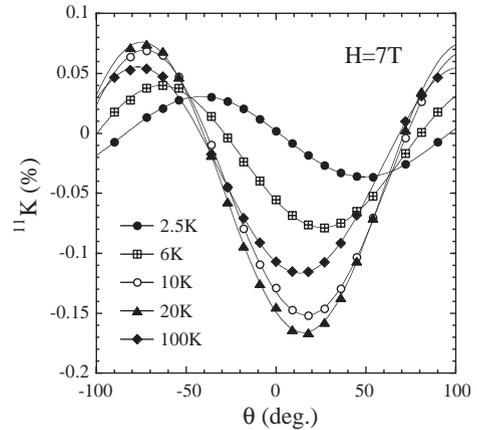


Fig. 2. Angular dependence of the shift <sup>11</sup>K of B(1) site obtained at various temperatures with the field of 7 T applied perpendicular to [1  $\bar{1}$  0].  $\theta$  is the angle between the field and the *c*-axis.

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