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Spin superstructure in the $\frac{1}{8}$-magnetization plateau phase of the 2D orthogonal dimer spin system $SrCu_2(BO_3)_2$

K. Kodama[a], M. Takigawa[a,*], M. Horvatić[b], C. Berthier[b,c], H. Kageyama[a], Y. Ueda[a], S. Miyahara[a,d], F. Becca[d], F. Mila[d]

[a] *Institute for Solid State Physics, University of Tokyo, 5-1-5 Kashiwanoha, 277-8581 Chiba, Japan*
[b] *Grenoble High Magnetic Field Laboratory, CNRS and MPI-FKF, Grenoble 38042, France*
[c] *Laboratorie de Spectrométrie Physique, Université Joseph Fourier Grenoble I, St.-Martin d'Hères 38402, France*
[d] *Institut de Physique Théorique, Université de Lausanne, Lausanne CH-1015, Switzerland*

Abstract

Nuclear magnetic resonance (NMR) experiments have been performed on the quasi-two-dimensional quantum spin system $SrCu_2(BO_3)_2$. The Cu NMR spectrum in the $\frac{1}{8}$-magnetization plateau state at 27.6 T and 35 mK shows fine structure, providing clear evidence for a magnetic superlattice in the plateau state. We conclude that the superlattice has a rhomboid unit cell with oscillating spin polarization, which is in good agreement with the numerical calculation on the Shastry–Sutherland spin model.

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Keywords: Quantum phase transition; Magnetization plateau; Spin superstructure

$SrCu_2(BO_3)_2$ has a two-dimensional network of orthogonal dimers of spin-$\frac{1}{2}$, which is equivalent to the Shastry–Sutherland spin model [1]. The ground state is the dimer singlet state [2] with an excitation gap [3] and the dispersion of the triplet excitations has an extremely small width [4], in accord with the prediction for the Shastry–Sutherland model [5]. The most striking property of this compound are the plateaus in the magnetization curve at $\frac{1}{8}$, $\frac{1}{4}$ and $\frac{1}{3}$ of the fully saturated moment [3,6]. Theoretical studies have suggested that the excited triplets with small kinetic energy localize due to repulsive interactions to form a superlattice [7], when the density of the triplets is commensurate with the crystal lattice. This is analogous to the Wigner crystallization or charge ordering in electron systems. The inequivalent Cu-sites in the superlattice should have different hyperfine fields, which can be observed by NMR. We have performed NMR measurements on a single crystal of $SrCu_2(BO_3)_2$ in the magnetic field up to 28 T applied along the *c*-axis at 35 mK [8]. This field range covers the $\frac{1}{8}$ plateau state, which is observed for 27–28.5 T.

Fig. 1 shows the Cu NMR spectra at 26 T (inset) and at 27.6 T (main panel). One type of Cu-sites with a specific value of the hyperfine field generally gives six NMR lines, three lines split by electric quadrupolar interaction for each of two isotopes ^{63}Cu and ^{65}Cu. The spectrum at 26 T can indeed be represented as a combination of such six lines, indicating that the magnetization is largely uniform at this field except for some degree of random distribution causing the finite line width.

At 27.6 T, the field near the middle of the $\frac{1}{8}$-magnetization plateau, we observed a completely different spectrum with a large number of sharp peaks distributed over a wide frequency range. This provides clear evidence for a magnetic superlattice with broken translational symmetry in the $\frac{1}{8}$ plateau phase. The six sharp peaks in the frequency range 105–165 MHz are ascribed to one type of Cu sites with the negative

*Corresponding author. Tel.: +81-4-7136-3225; fax.: +81-4-7136-3225.
E-mail address: masashi@issp.u-tokyo.ac.jp (M. Takigawa).

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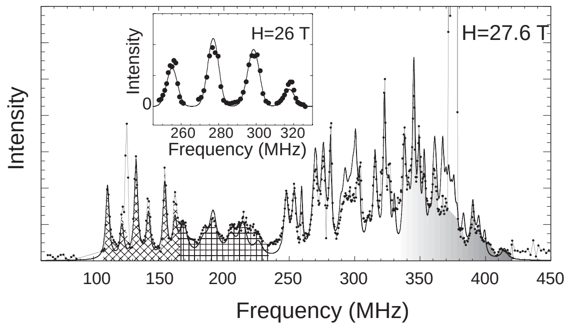

Fig. 1. Cu NMR spectra obtained at $T = 35$ mK is shown by black dots. The inset is the spectrum at $H_0 = 26$ T, and the main panel is the one at $H_0 = 27.6$ T.

hyperfine field $H_n = -16.2$ T, corresponding to the largest local magnetization $\langle S_z \rangle = 0.30$. Similarly, the broad peaks for 165–235 MHz represent another type of sites with $\langle S_z \rangle = 0.20$. The rest of the spectrum comes from the sites with much smaller values of $\langle S_z \rangle$. It should be noted, however, that some of these sites have positive hyperfine fields, i.e., spins polarized opposite to the field. Thus the magnetization oscillates within the unit cell of the superlattice.

We found that any satisfactory fitting of the spectrum requires at least 11 distinct Cu sites. This fact leads to the conclusion that the superlattice has the rhomboid unit cell as shown in Fig. 2, since any other types of unit cell cannot produce such a large number of inequivalent sites. The number of inequivalent Cu sites for the rhomboid cell is eight for a single layer. However, it may increase up to 16 depending on the pattern of stacking the layers along the c-axis. The magnetization profile calculated by the exact diagonalization of the Shastry–Sutherland model on a 16-spin cluster of a rhomboid cell [8,9] is shown in Fig. 2. One can clearly recognize a building unit consisting of one strongly polarized dimer surrounded by decaying oscillation of magnetization. It resembles the Friedel oscillation near impurities in metals. The distribution of hyperfine field at Cu site derived from the calculated magnetization profile captures the essential feature of the experimental results, indicating that such a magnetic superstructure is indeed realized in the $\frac{1}{8}$ plateau phase.

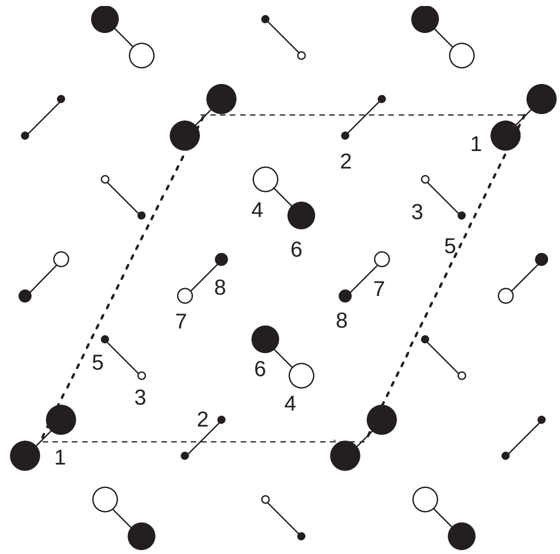

Fig. 2. Magnetization profile in the $\frac{1}{8}$ plateau state. Closed (open) cirle shows the positive (negative) $\langle S_z \rangle$. The circle size represents the magnitude of $\langle S_z \rangle$.

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