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1/3 Magnetization Plateau in $SrCu_2(BO_3)_2$ - Stripe Order of Excited Triplets -

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High-field magnetization measurements up to 57 T have been performed at 0.08 K in a single crystal of a two-dimensional spin-gap material $SrCu_2(BO_3)_2$. We successfully observed the predicted plateau at 1/3 of the total magnetization around 50 T, in which the magnetic superstructure is characterized by a novel stripe order of triplets. The 1/3 plateau is much wider than the previously observed 1/4 and 1/8 plateaux.

KEYWORDS: $SrCu_2(BO_3)_2$, Shastry-Sutherland model, quantized magnetization plateaux, stripe, spin gap

There has been a resurgent interest in the phenomena of quantized magnetization plateaux because the plateaux of recent interest are caused principally by quantum many-body effects,[1)] in contrast to classical systems previously investigated such as $CoCl_2 2H_2O$.[2)] Experimentally, several quantum-spin systems were reported to have magnetization plateaux. A 1/2 plateau of the total magnetization was observed in $[Ni_2(Medpt)_2(\mu\text{-ox})(\mu\text{-}N_3)]ClO_4 0.5H_2O$,[3)] 1/4 and 3/4 in NH_4CuCl_3,[4)] and 1/8 and 1/4 in $SrCu_2(BO_3)_2$.[5)]

$SrCu_2(BO_3)_2$ is a layered material with a tetragonal symmetry, composed of alternating stacks of $2CuBO_3$ and Sr planes along the c axis. In the $CuBO_3$ plane, nearest-neighbor Cu^{2+} ions constitute an $S = 1/2$ dimeric unit, and each dimer connects orthogonally with four nearest-neighbor dimers. The intradimer and interdimer interactions, symbolized by J and J', respectively, lead to an exact ground state with spin gap Δ, thus realizing the 2D Shastry-Sutherland model.[6,7)] The values of J, J' and Δ were estimated to be 100 K, 68 K and 34 K, respectively.[8-10)]

The 1/4 and 1/8 plateaux in $SrCu_2(BO_3)_2$ were first detected by magnetization measurements on a polycrystalline sample performed in a magnetic field H up to $H = 43$ T and down to 1.5 K with a pulse magnet at ISSP, the University of Tokyo,[5)] followed by those using field-oriented samples for $H /\!/ c$ and $H \perp c$[10)] and a single crystal for $H \perp c$.[11)] Theoretically, Miyahara and Ueda explained that the origin of the plateaux lies in the almost localized nature of the triplet excitations.[7)] Moreover, in light of the tetragonal symmetry of the crystal structure, they initially argued the following necessary condition for the triplets to ensure ordered rather than disordered structures: *corresponding magnetic superstructures should also be tetragonal symmetry.* In fact, this condition satisfies the superstructures shown in Figs. 1(a) and 1(b) for the 1/8 and 1/4 plateaux, respectively. Following this simple geometrical argument, one expects other plateaux at 1/2, 1/10, 1/16, 1/32..., which also have *square* unit cells.

However, recent detailed analyses carried out independently by Momoi and Totsuka[11)] and by Miyahara and Ueda[12)] have revealed the possibility that the superstructure for the 1/4 plateau is of *stripe* type with a *rectangular* unit cell, as shown in Fig. 1(c). Novel anisotropic interactions cause the stripe order. Namely, the interaction of the next-nearest-neighbor pair of triplets is much weaker than that of the third-nearest-neighbor one. By the same logic, a prediction has been made that a 1/3 plateau with a superstructure of *stripe* order (see Fig. 1(d)) will be observable if a magnetic field higher than that ever studied is applied.

It is unfortunate that, in spite of intensive experimental activities,[5, 8-10, 13-15)] the existence of the stripe order even for the 1/4 plateau has yet not been confirmed. This is partly because the required magnetic field is beyond the capability of facilities currently used for microscopic techniques such as neutron diffraction. In this letter, we measured the magnetization at 0.08 K for single-crystalline $SrCu_2(BO_3)_2$ up to 57 T to successfully observe the 1/3 plateau. It should be emphasized that, unlike the 1/4 plateau, the stripe superstructure (Fig. 1(d)) is the *unique* candidate for the 1/3 plateau.

The high-field magnetization measurements were conducted using pulse magnets at the High Magnetic Field Laboratory, KYOKUGEN in Osaka University. High-quality single crystals of $SrCu_2(BO_3)_2$ were obtained by a travelling solvent floating-zone technique, as described in ref. 16. We used one of the grown single crystals of high quality with the approximate dimensions of 2 mm×2 mm×2 mm. Magnetization data for $H \perp c$ were collected at 0.08 K in magnetic fields up to 57 T, and

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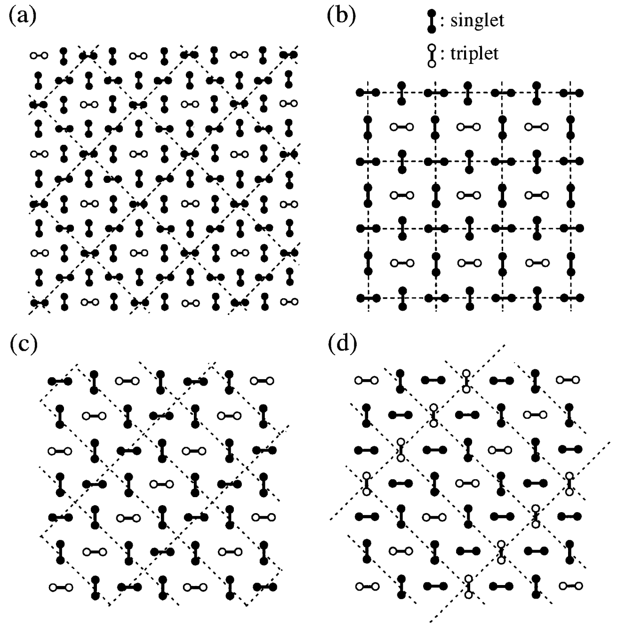

Fig. 1. Possible ordered structures between the excited triplets (open) and the remaining singlets (solid) for the 1/8 plateau (a) the 1/4 plateau (b) and (c), and the 1/3 plateau (d). The magnetic unit cells are represented by broken lines.

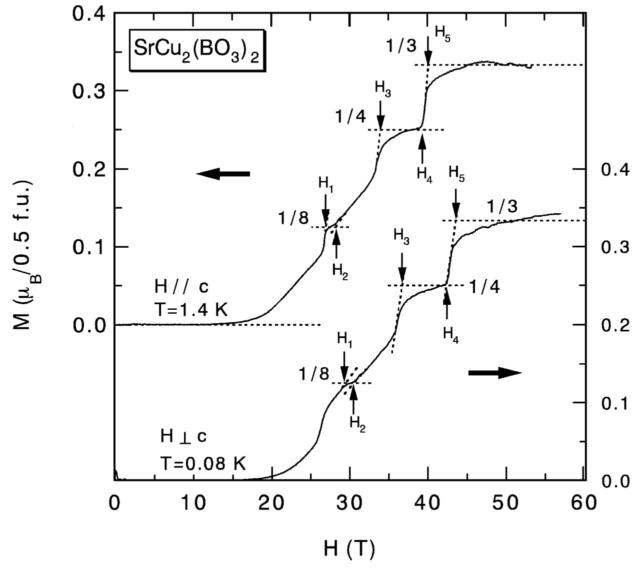

Fig. 2. The magnetization curves measured at 0.08 K for $H \perp c$ and at 1.4 K for $H/\!/c$. The broken lines correspond to 1/3, 1/4 and 1/8 of the total magnetization.

those for $H/\!/c$ were collected at 1.4 K up to 53 T.

Figure 2 shows the magnetization curves for $H/\!/c$ and for $H \perp c$, where within the experimental accuracy, no hysteresis was observed upon increasing or decreasing the external field. It appears that the difference between the two curves arises only from the g-factor. As seen in Fig. 3, the parallel and perpendicular magnetizations can be normalized using $g_{/\!/} = 2.28$ and $g_{\perp} = 2.05$ determined by electron spin resonance (ESR).[10] Strictly speaking, there is a slight difference around 30 T, which will be discussed later. Note that this is the first measurement of parallel magnetization using the single crystal. The obtained magnetization curve for $H/\!/c$ has more distinct

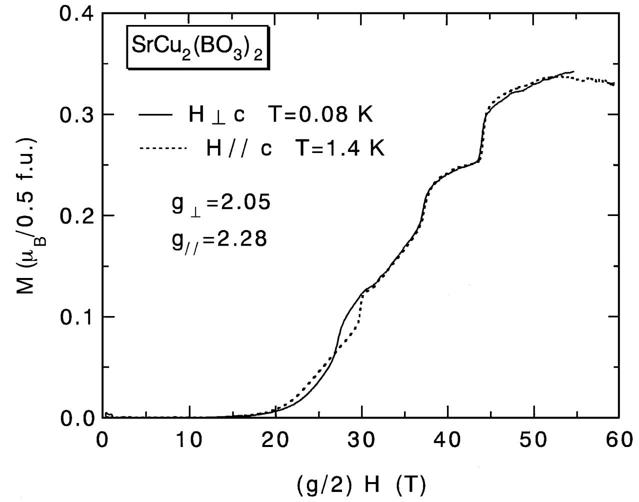

Fig. 3. Normalized parallel and perpendicular magnetizations for $H \perp c$ (solid) and for $H/\!/c$ (broken), where $g_{/\!/} = 2.28$ and $g_{\perp} = 2.05$ are used.

1/4 and 1/8 plateaux compared with that obtained using the field-oriented polycrystal[10] with poor orientation, as evaluated by nuclear magnetic resonance. On the other hand, the result for $H \perp c$ below 42 T is almost identical to that previously reported using the single crystal.[9] As shown in Fig. 2, the critical fields are determined to be $H_1 = 29.6\,\mathrm{T}$, $H_2 = 31.2\,\mathrm{T}$, $H_3 = 36.7\,\mathrm{T}$ and $H_4 = 43.6\,\mathrm{T}$ for $H \perp c$, and $H_1 = 26.9\,\mathrm{T}$, $H_2 = 28.2\,\mathrm{T}$, $H_3 = 34.1\,\mathrm{T}$ and $H_4 = 39.1\,\mathrm{T}$ for $H/\!/c$, which is almost consistent with the previous results.[9,10]

A glance at Fig. 2 reveals a wide plateau at 1/3 of the full magnetization. As mentioned earlier, the 1/3 plateau cannot accommodate any magnetic superstructure with a square unit cell. Therefore, this observation confirms, for the first time, the existence of the novel stripe order, where the magnetic unit cell is composed of four singlets and two triplets. The onset field H_5 of the 1/3 plateau is estimated to be 43.6 T for $H \perp c$ and 40.2 T for $H/\!/c$. As evident in Fig. 2, the 1/3-plateau phase is stable over a much wider field range than the 1/8 and 1/4 plateaux. Another interesting point is the sudden jump of the magnetization between the 1/4 and 1/3 plateaux, possibly indicating a first-order phase transition, although there is no hysteresis between the field increase and decrease processes.

There are several theoretical papers concerning calculations of the magnetizations with varying J'/J.[11,12,17] In Fig. 4, we compare the experimental curve with the calculations in terms of a hard-core boson model by Momoi and Totsuka[11] (theory I) and Miyahara and Ueda[12] (theory II), where $J = 100\,\mathrm{K}$ and $J' = 68\,\mathrm{K}$ are used. It is noteworthy that both models quantitatively reproduce the experimental data, particularly around the 1/3 plateau. The main difference between theories I and II, in short, is that the former considers correlated hopping terms and neglects long-range interactions, whereas the case is reversed in the latter. In connection with this difference, the 1/3 plateau ends at 80 T according to theory I, while theory II predicts that it ends at 110 T. Moreover, theory II concluded that a 1/2 plateau appears, whereas theory I did not. Thus, the future direction of

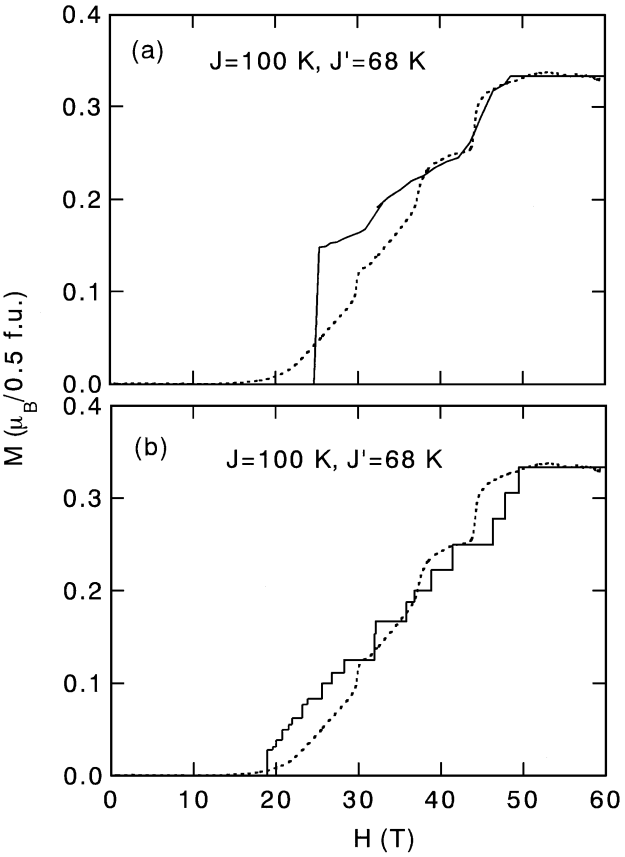

Fig. 4. Comparison with the theories by Momoi and Totsuka[11] (a) and Miyahara and Ueda[12] (b), where $J = 100\,\mathrm{K}$ and $J' = 68\,\mathrm{K}$ are assumed. The dotted curve is the experimental result for $H /\!/ c$ normalized by $g = 2.28$.

this area of study will be one that requires even higher magnetic fields.

Finally, let us discuss the anomalous behavior around 30 T, as shown in Fig. 3. In ref. 9, we suggested the 1/10 plateau, judging from the perpendicular magnetization that displays a bump at about 1/10 of the total magnetization, as observed in this experiment as well. The behavior of the parallel magnetization, however, is not indicative of the 1/10 plateau, as seen in Fig. 3. It is possible that the appearance of the 1/10 plateau depends on the field direction, i.e., the orientation of the excited triplets. Alternatively, this phenomenon could be a quantum effect of some kind.

In conclusion, our high-field magnetization measurement for $SrCu_2(BO_3)_2$ allowed the first experimental verification of the stripe order in the orthogonal dimer lattice through the observation of the 1/3 plateau that appears around 50 T. The transition between the 1/3 and 1/4 plateaux seems to be of the first order. The calculated magnetizations satisfactorily reproduce the experimental magnetization result around the 1/3 plateau region.

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