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# High field properties of the frustrated 2D dimer spin system $\text{SrCu}_2(\text{BO}_3)_2$

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**Abstract.** We discuss high-field magnetic properties of the quasi two dimensional frustrated dimer spin system  $\text{SrCu}_2(\text{BO}_3)_2$ , which is a realization of the Shastry-Sutherland spin model and shows magnetization plateaus at  $1/8$ ,  $1/4$  and  $1/3$  of the saturated magnetization. Based on the results of  $^{11}\text{B}$  NMR experiments, we discuss (1) the spin superstructure in the plateau phases, (2) the effects of anisotropic interactions such as the Dzyaloshinski-Moriya interaction, and (3) nature of the phases above the  $1/8$  plateau.

## 1. Introduction

Properties of coupled singlet dimer spin systems in high magnetic field have been attracting strong recent attention [1]. At zero-field, the ground state of these materials is simply a collection of singlet dimers. The low lying excitations are usually described by  $S=1$  triplet magnons with an energy dispersion  $E(\mathbf{q})$  as a function of momentum  $\mathbf{q}$ . The width of the dispersion is determined by the inter-dimer exchange interactions and the minimum energy of the dispersion defines the spin-gap  $\Delta$ . A magnetic field applied to such a system causes the Zeeman splitting of the triplets, reducing the gap to the lowest  $S_z = 1$  branch as  $\Delta - g\mu_B H$ . This gap vanishes at the critical field  $H_{c1} = \Delta/(g\mu_B)$ . A fascinating aspect of the field-induced quantum phase transition in coupled dimer systems is the rich variety of phenomena encountered in various materials, even though the systems are conceptually very simple.

For the fields near  $H_{c1}$ , each dimer has two low energy states. the singlet and the  $S_z = 1$  triplet. They may be described by up or down states of a effective spin  $1/2$ , which in turn can be mapped onto a hard core boson model. The magnetic field sets the chemical potential for bosons. For the field above  $H_{c1}$ , there are finite density of bosons at  $T=0$ , which should

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undergo Bose-Einstein condensation (BEC). In the original spin model, this corresponds to a coherent superposition of the singlet and the triplet states. As a consequence, both the uniform magnetization parallel to the field and the staggered (AF) magnetization perpendicular to the field appear above  $H_{c1}$  [2, 3]. Although field-induced AF transitions have been observed in a number of materials such as  $\text{TlCuCl}_3$  [4], the BEC description is strictly valid only when the interactions are isotropic in the plane perpendicular to the field so that the phase of the staggered magnetization can take any value. In real materials, however, there are always small anisotropic perturbations. They may significantly affect the nature of the phase transition, or even completely eliminate the phase transition, as we discuss later.

In many cases, the magnetization increases smoothly above  $H_{c1}$  up to saturation with increasing magnetic field. In some materials with frustrated interactions, however, the magnetization stays constant at fractional values of the saturated magnetization for a finite range of magnetic field [5]. This means that the density of bosons is kept constant against the change of chemical potential, indicating opening of a gap in the excitation spectrum. Oshikawa *et al.* [6, 7] developed a topological theory and derived a general condition for the plateau formation. It follows that if a magnetization plateau appears at a fractional value  $S - m = p/q$  ( $p$  and  $q$  are mutually prime) with  $q$  greater than the number of sites in a crystalline unit cell, the ground state must break the translational symmetry of the crystalline lattice. Such a superstructure should be a consequence of localization of triplets due to repulsive interactions.

In common with other interacting quantum systems, the balance between the kinetic energy and the interaction energy determines whether the particles are localized or itinerant. For the coupled dimers, the repulsive interaction is given by the  $z$ -component (parallel to the field) of the interdimer interactions, while the kinetic energy is determined by their  $xy$ -components. If the interactions are frustrated, the kinetic energy is generally suppressed and magnetization plateaus may appear. The nature of the phases in the neighborhood of plateaus are particularly interesting since they are analogous to doped Mott insulators. At the edges of the plateaus, quantum melting of the triplet lattice leading to a insulator to superfluid transition may be expected. In some cases, however, triplet lattice may persist outside the plateau while the doped triplet condense into a superfluid component. Since such a supersolid phase has been proposed numerically for certain lattice-boson models [8], it is challenging to find it in real materials.

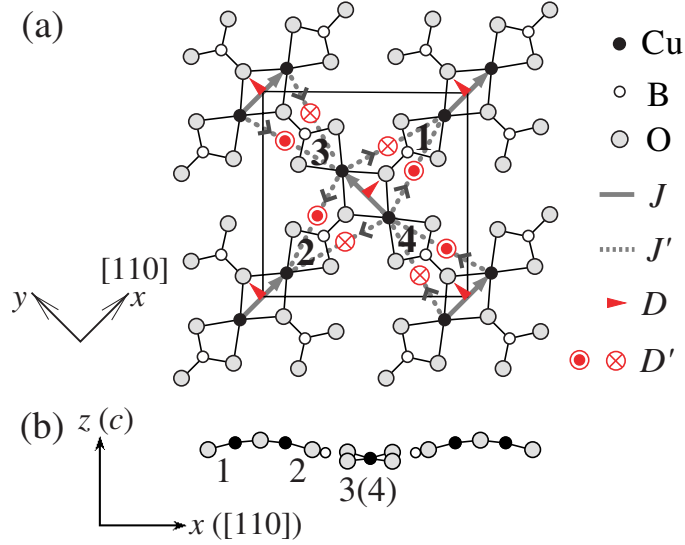
The first direct observation of symmetry breaking magnetic superstructure associated with a magnetization plateau was reported for  $\text{SrCu}_2(\text{BO}_3)_2$ , a quasi 2D coupled dimer spin system, by Kodama *et al.* [9] based on nuclear magnetic resonance (NMR) experiments at Cu sites. In this paper, we review the present understanding of the high field properties of  $\text{SrCu}_2(\text{BO}_3)_2$ .

## 2. Magnetization plateaus in $\text{SrCu}_2(\text{BO}_3)_2$

The crystal structure of  $\text{SrCu}_2(\text{BO}_3)_2$  consists of magnetic  $\text{CuBO}_3$  layers and non-magnetic Sr layers alternating along the  $c$ -axis. The magnetic layers shown in Fig. 1 contains orthogonal dimers formed by pairs of  $\text{Cu}^{2+}$  ions each carrying spin 1/2. The simplest spin model appropriate for this material is the Shastry-Sutherland model [10, 11, 12]

$$\mathcal{H}_0 = J \sum_{n.n.} \mathbf{s}_i \cdot \mathbf{s}_j + J' \sum_{n.n.n.} \mathbf{s}_i \cdot \mathbf{s}_j. \quad (1)$$

where  $J$  and  $J'$  are the nearest neighbor (intra-dimer) and next-nearest neighbor (interdimer) isotropic exchange interactions. The ground state of this model is obvious in the two limiting cases: when  $J/J' \gg 1$ , it reduces to a collection of dimer singlets, while if  $J/J' \ll 1$ , the model is equivalent to an antiferromagnet on a square lattice, hence, the ground state has a Néel order. Whether there are other phases between the two limits is still an open question. It is known that for  $J/J' < 0.68$  the simple product of dimer singlets is the *exact* ground state. Various



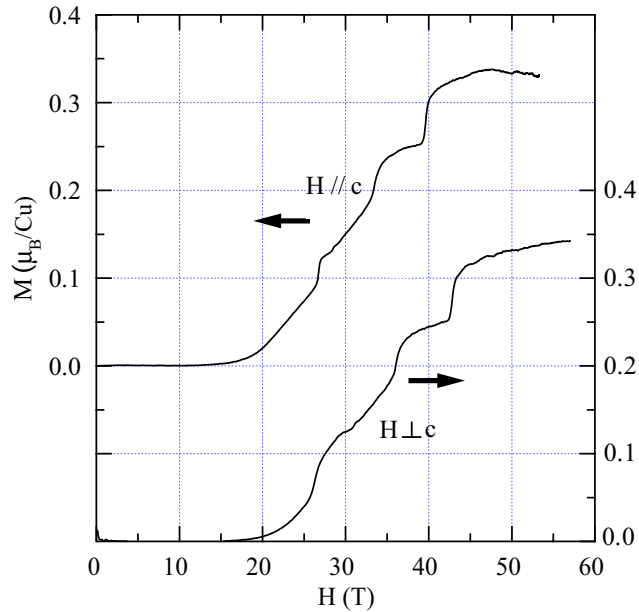
**Figure 1.** The magnetic layer of  $\text{SrCu}_2(\text{BO}_3)_2$  viewed along (a) the  $c$ -axis and (b) the  $[\bar{1}\bar{1}0]$ -directions. Numbers distinguish different Cu and B sites in a unit cell. Symbols for  $D$  and  $D'$  indicate the direction of  $\mathbf{d}$ -vector in the Dzyaloshinski-Moriya interaction expressed as  $\mathbf{d} \cdot (\mathbf{s}_i \times \mathbf{s}_j)$  with the bond direction  $i \rightarrow j$  shown by arrows.

experiments have established that  $\text{SrCu}_2(\text{BO}_3)_2$  has the dimer-singlet ground state with the spin-gap  $\Delta = 33$  K at zero magnetic field. The values of the exchange parameters were estimated as  $J/J' = 0.64$  and  $J = 85$  K from fits to the magnetic susceptibility [12]. Slightly different values  $J/J' = 0.60$  and  $J = 71$  K were obtained from the analysis of the excitation spectra [13].

It is known that the frustration between  $J$  and  $J'$  of the Shastry-Sutherland model strongly suppresses the kinetic energies of triplets [11]. Early neutron scattering experiments [14] indeed showed extremely small dispersion width of single triplet excitations in  $\text{SrCu}_2(\text{BO}_3)_2$ . Subsequent experiments with higher resolution [15, 16] demonstrated that even this small dispersion is due to the secondary interdimer Dzyaloshinski-Moriya interaction between next nearest neighbors ( $D'$  in Fig. 1), which is not included in  $\mathcal{H}_0$ .

The most striking property of  $\text{SrCu}_2(\text{BO}_3)_2$  is the magnetization plateaus at  $1/8$ ,  $1/4$ , and  $1/3$  of the full saturation shown in Fig. 2 [17]. From the commensurability condition mentioned above, the ground states at these plateaus are expected to break the translational symmetry due to formation of the superlattices of triplets. For the  $1/8$  plateau, symmetry breaking spin superstructure has been indeed observed by nuclear magnetic resonance (NMR) experiments at the Cu and B nuclei performed with the 20MW resistive magnet at the Grenoble High Magnetic Field Laboratory [9, 18].

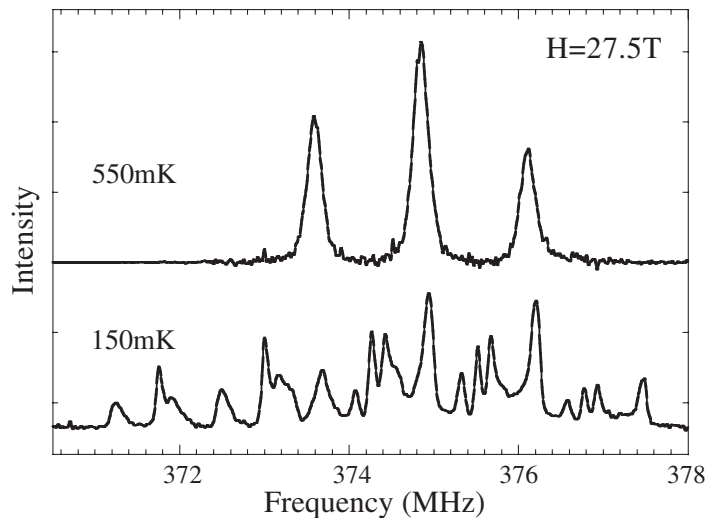
Figure 3 shows the  $^{11}\text{B}$  NMR spectra for the field of 27.5 T at two different temperatures. At high temperatures, the spectrum shows three resonance lines split by nuclear quadrupole interaction. This indicates that all the nuclei experience a unique magnetic hyperfine field, therefore, the magnetization is uniform. The spectrum at lower temperature, on the other hand, develop a large number of sharp peaks. This spectrum can be fit by convoluting the quadrupole split three lines with properly chosen distribution of the magnetic hyperfine field. We found that the distribution of the hyperfine field can be reproduced well by the spin-density profile shown in the left panel of Fig. 4 [18, 19], obtained from the exact diagonalization of the Shastry-Sutherland model with additional spin-lattice coupling [20]. Since the calculated result was shown to be compatible with the Cu NMR spectrum in the  $1/8$  plateau phase [9], the B



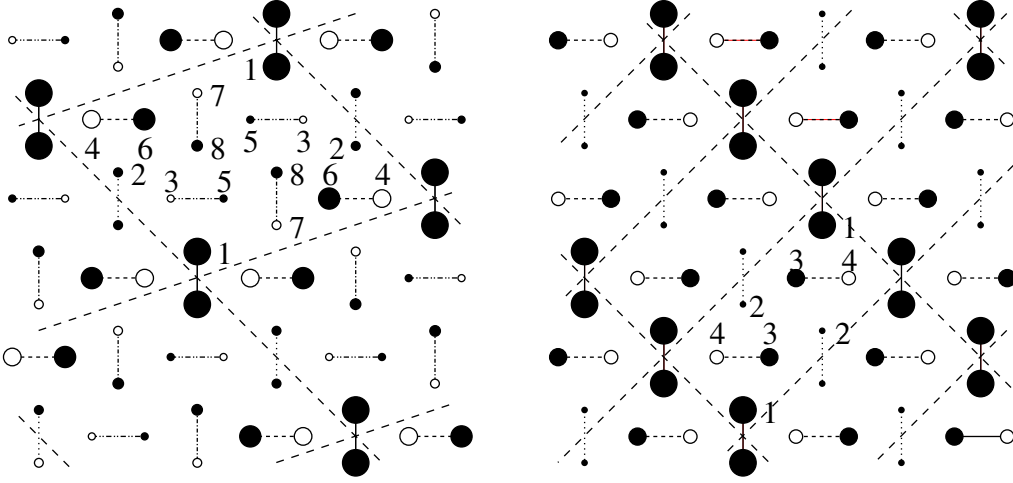
**Figure 2.** The field dependence of the magnetization in  $\text{SrCu}_2(\text{BO}_3)_2$  for two different field directions (from Ref. [17]).

NMR results provide further conclusive support for the spin structure shown in Fig. 4.

A prominent feature of the calculated profile in the  $1/8$  plateau is that a triplet is not confined on a single dimer but spread over three dimers surrounded further by small oscillating spin density. The negative polarization (opposite to the field direction) on site 4 is due to unfrustrated antiferromagnetic exchange field from the nearest neighbor dimer (site 1) which is nearly fully polarized. The existence of two sites with large positive polarization and one with large negative polarization has been unambiguously confirmed from the Cu and B NMR spectra. It should be noted that such longitudinal staggered magnetization within one dimer require mixing of the singlet and the  $S_z = 0$  triplet states, therefore, cannot be obtained by effective



**Figure 3.** The  $^{11}\text{B}$  NMR spectra for the field of approximately 27.5 T.



**Figure 4.** The spin density profile for the 1/8 plateau with the rhomboid supercell (left panel) and for the 1/4 plateau (right panel) obtained from exact diagonalization of the Shastry-Sutherland model (from Ref. [20]).

models of spin 1/2 or one component bosons. Similar "three dimer" structure of triplets appear also in the calculated profiles for the 1/4 plateau (the right panel of Fig. 4) and the 1/3 plateau (not shown).

### 3. Effects of anisotropic interactions

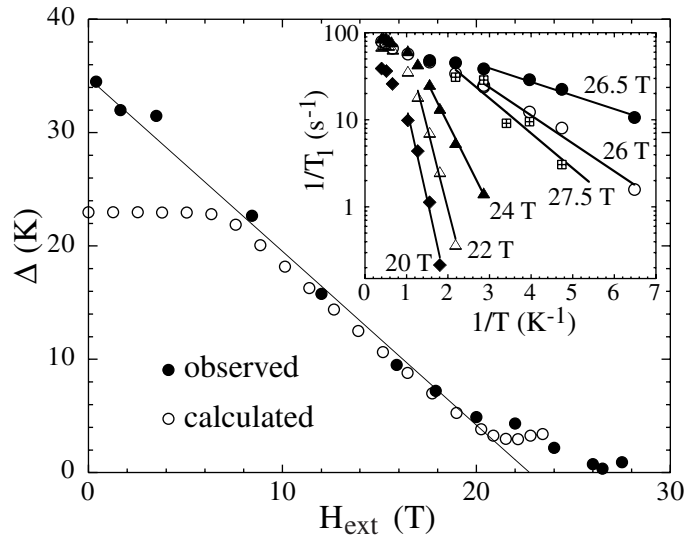
The magnetization data in Fig.2 has a peculiar feature. From the value of the gap at zero-field (33 K) and the  $g$ -values ( $g_c=2.28$ ,  $g_a=2.05$  [21]), one would expect the gap to close at  $H_{c1}=21.5$  T (24 T) for the field along (perpendicular to) the  $c$ -direction and the BEC phase to appear above  $H_{c1}$ . On the contrary, the magnetization starts to rise gradually around 15 T (19 T) for  $H \parallel c$  ( $H \perp c$ ), which is much smaller than  $H_{c1}$ , and there is no signature for a phase transition up to the boundary to the 1/8 plateau. The EPR [21, 22] and the specific heat [23] data also indicated that the gap does not close at  $H_{c1}$ . These results suggest existence of anisotropic interactions, which mix the singlet and the  $S_z=1$  triplet states. In fact, non-coplanar buckling of the magnetic layers (Fig. 1) violating inversion symmetry at the center of dimer bonds allows the intra-dimer Dzyaloshinski-Moriya interaction (indicated by  $D$  in Fig. 1) and the staggered  $g$ -tensor of the following form,

$$\mathcal{H}_1 = -\mu_B \mathbf{H} \cdot \left( \sum_{i=1}^4 \mathbf{g}_i \cdot \mathbf{s}_i \right) + D \left\{ \sum_A (s_1^z s_2^x - s_1^x s_2^z) - \sum_B (s_3^y s_4^z - s_3^z s_4^y) \right\}, \quad (2)$$

where A (B) denotes dimers along the  $x$ - ( $y$ ) direction and  $\mathbf{s}_1 \sim \mathbf{s}_4$  are the Cu spins at four different sites in the unit cell (Fig. 1). The  $g$ -tensor is given by

$$\mathbf{g}_1 = \begin{pmatrix} g_x & 0 & -g_s \\ 0 & g_y & 0 \\ -g_s & 0 & g_z \end{pmatrix},$$

for site 1, while  $\mathbf{g}_2$ ,  $\mathbf{g}_3$ , and  $\mathbf{g}_4$  are obtained from  $\mathbf{g}_1$  by symmetry operations of the crystal. The sign of the  $xz$ -component is opposite for  $\mathbf{g}_2$ , i.e.,  $g_s$  represents the staggered component.



**Figure 5.** Field dependence of the excitation gap (solid circles in the main panel) determined from the activation energy of  $1/T_1$  (inset) are compared with the results of exact diagonalization calculation (open circles). The constant behavior of the calculated gap at low fields is due to the singlet bound state of two triplets, which has lower energy than one triplet but does not contribute to the nuclear relaxation rate.

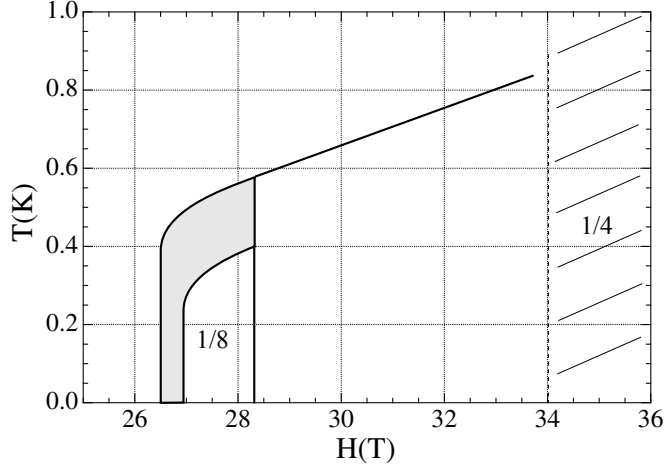
It should be noted that the effects of the intra- and inter-dimer DM interactions are completely different: the former mixes the singlet and the triplet states on the same dimer, while the latter promotes hopping of the triplets between neighboring dimers.

The anisotropic interactions in Eq. 2 result in non-collinear staggered magnetization consisting of four sublattices. The field-induced staggered magnetization has been indeed detected by measuring the orientation dependence of the NMR frequency shift at B and Cu sites [24, 25]. Estimating  $g_s=0.023$  from the anisotropy of the diagonal components of the  $g$ -tensor and the structural data, the value of  $D$  was determined as  $D/J=0.034$  from our NMR data. This result is in good agreement with the independent estimates based on EPR [26] and optical absorption [27] experiments. The NMR results indicate sizable staggered moment of about  $0.1 \mu_B$  at low temperature near  $H_{c1}$ . This was reproduced by numerical calculation for the Hamiltonian of Eq. 2 on a small cluster [24]. The absence of BEC transition at  $H_{c1}$  can be now understood, since the direction of the field-induced staggered magnetization is fixed uniquely, already breaking the rotational symmetry perpendicular to the field direction before the BEC transition could occur. We expect that the staggered magnetization remains to be large in the  $1/8$  plateau and higher field phases. However, no experimental information is yet available for this field range.

The singlet-triplet mixing induced by the anisotropic interactions causes level anti-crossing near  $H_{c1}$  as observed by EPR [21, 22]. The persistence of the gap near  $H_{c1}$  was also evidenced by the nuclear relaxation data as shown in Fig. 5. An interesting feature is that the gap nearly vanishes at 26.5 T, which is the boundary to the  $1/8$  plateau phase. The nearly vanishing gap at the edges of the plateau phase has been also found in a recent numerical studies on frustrated spin ladders [28].

#### 4. Above the $1/8$ plateau

We have recently performed  $^{11}\text{B}$  NMR experiments in higher fields up to 31 T applied along the  $c$ -axis to explore the phases above the  $1/8$  plateau. While the spectral shape is identical



**Figure 6.** Schematic phase diagram of  $\text{SrCu}_2(\text{BO}_3)_2$  in high magnetic field.

within the  $1/8$  plateau phase, a sudden change occurred at 28.4 T, which is nearly independent of temperature. This field is identified as the upper boundary of the  $1/8$  plateau, in agreement with the magnetization data. The spectral shape changes continuously with field and temperature in the phase above the plateau. In spite of the change in the spectral shape, the overall range of the distribution of magnetic hyperfine field remains unchanged from that in the  $1/8$  plateau (lower panel of Fig. 3). From the analysis of the spectrum in the  $1/8$  plateau, we assigned the peak at the lowest frequency to the B sites next to nearly fully polarized dimers [18]. Persistence of NMR lines with such a large negative hyperfine field, therefore, indicates that nearly fully polarized dimers still exist in the higher field phase. In other words, the superlattice of the triplet survives above the plateau.

With increasing temperature, the wide distribution of hyperfine field disappears discontinuously, resulting in quadrupole split sharp NMR lines. This transition is similar to what was observed in the  $1/8$  plateau (Fig. 3) but occurs in a much narrower temperature range. From a series of NMR measurements, the phase diagram can be constructed as schematically shown in Fig. 6. All the phase boundaries appear to be first order. The transition temperature between the paramagnetic and the ordered phases increases smoothly with increasing temperature across the phase boundary. Such behavior is quite different from the "dome" like phase diagram observed for a typical field-induced BEC phase in systems without plateaus. This transition has been originally discovered by Tsujii *et al.* by specific heat measurements up to 33 T [23]. Their data for the transition temperature smoothly extrapolates to our result. We now identify the transition as the solidification (superlattice formation) of triplet breaking the translational symmetry, analogous to the transition into the plateau phase.

## 5. Summary

The unique properties of  $\text{SrCu}_2(\text{BO}_3)_2$  revealed by high field NMR experiments are reviewed. The intradimer DM interaction and the staggered  $g$ -tensor play important roles to eliminate the BEC transition and induce sizable staggered magnetization. Symmetry breaking superlattice of the triplets survives in the intermediate phase between the  $1/8$  and the  $1/4$  phases with smooth variation of the transition temperature.

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