Field-Induced Phase Transitions Driven by Quantum Fluctuation in $S = 1/2$ Anisotropic Triangular Antiferromagnet Cs$_2$CuBr$_4$

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The field induced magnetic phase transitions of Cs$_2$CuBr$_4$ were investigated by means of magnetization process and neutron scattering experiments. Cs$_2$CuBr$_4$ should be characterized as $S = 1/2$ two-dimensional triangular antiferromagnet. Below the ordering temperature $T_N = 1.4$ K, the spin structure is the helical incommensurate structure almost within the triangular lattice plane. In the field direction within the triangular lattice plane, Cs$_2$CuBr$_4$ exhibits the magnetization plateaux at one-third and two-thirds of the saturation magnetization. The spin structure in the one-third plateau phase is found to be almost collinear $up-up-down$ structure which should be stabilized by quantum fluctuation as predicted by the theoretical studies.

§1. Introduction

Triangular antiferromagnets (TAF) have been of great interest from the viewpoint of the interplay of spin frustration and quantum fluctuation.\(^1\)–\(^3\) In classical Heisenberg TAF, the ground state has continuous degeneracy even in the magnetic field due to the spin frustration. For this case, there is no phase transition occurs up to saturation, so that the magnetization curve is monotonic. For quantum Heisenberg TAF with small spin $S$, the quantum fluctuation plays an important role in determining the ground state, because the quantum fluctuation can remove the continuous degeneracy of the classical ground state. The quantum fluctuation in TAF

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was discussed theoretically using the spin wave theory\textsuperscript{1,3} and the numerical calculations\textsuperscript{4,5}. According to the theoretical studies, the up-up-down (\textit{uud}) spin structure is stabilized by the quantum fluctuation around the field at which the magnetization reaches one-third of the saturation magnetization $M_s$ for $S = 1/2$ two-dimensional (2D) Heisenberg TAF and exhibits the magnetization plateau.

The magnetization plateau is a new macroscopic quantum phenomenon corresponding to the quantization of magnetization. Recently, we found the quantum-fluctuation-assisted plateau at $M_s/3$ in Cs$_2$CuBr$_4$. Cs$_2$CuBr$_4$ has an orthorhombic crystal structure with space group $Pnma$.\textsuperscript{6} Figure 1(a) shows the crystal structure of Cs$_2$CuBr$_4$. The structure is composed of CuBr$_2^{-}$ tetrahedra and Cs$^+$ ions. The tetrahedra are linked along the $b$-axis. Cu$^{2+}$ ions with $S = 1/2$ form a distorted triangular lattice in the $bc$-plane as shown in Fig. 1(b).

The magnetic properties of isostructural Cs$_2$CuCl$_4$ have been extensively investigated by neutron scattering experiments.\textsuperscript{7–10} Cs$_2$CuCl$_4$ exhibits the helical magnetic ordering almost within the $bc$-plane (see Fig. 1(b)) below the ordering temperature $T_N = 0.62$ K with the ordering vector $q = (0, 0.528, 0)$. The incommensurate spin structure arises from the spin frustration between antiferromagnetic exchange interactions $J_1$ and $J_2$. From the results of inelastic neutron scattering, it was found that the exchange interactions $J_1$ and $J_2$ of Cs$_2$CuCl$_4$ are dominant and the inter layer coupling $J'$ is $J' \approx 0.05J_1$. Thus, Cs$_2$CuCl$_4$ was characterized as a quasi-2D frustrated spin system. Since Cs$_2$CuBr$_4$ has the same crystal structure as Cs$_2$CuCl$_4$, Cs$_2$CuBr$_4$ should also be described as quasi-2D distorted triangular system. We can expect a higher Néel temperature than in Cs$_2$CuCl$_4$, since the exchange interaction through bromine ions is generally stronger than that through chlorine ions. As described below, we present the results of magnetization process which exhibits the quantum-fluctuation-assisted plateaux and the preliminary result

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Fig. 1. (a) Perspective view of the crystal structure of Cs$_2$CuBr$_4$. (b) Antiferromagnetic interactions within the $bc$-plane and the helical incommensurate structure established in the ordered phase.
§2. Experiments

Single crystals of Cs$_2$CuBr$_4$ were grown by the slow evaporation method from the aqueous solution of CsBr and CuBr$_2$ in the mole ratio 2 : 1. The experimental details for pulsed high field magnetization process and the elastic neutron scattering experiments are described in the previous paper.$^{11}$ Inelastic neutron scattering experiments were performed at IN5 time-of-flight spectrometer of Institute Laue-Langevin, Grenoble. The sample used for this measurement was approximately 25 g in mass. All neutron scattering experiments were performed at $T \sim 50$ mK which was well below the ordering temperature $T_N = 1.4$ K using the dilution refrigerator.

§3. Results and discussion

Figure 2(a) shows the magnetization curve and its field derivative $dM/dH$ versus $H$ measured at $T \sim 400$ mK for $H \parallel c$ (whole results are reported on the previous paper$^{11}$). At the saturation fields ($H_s$) summarized in Table I, the value of the magnetization ($M_s$) is slightly larger than 1 $\mu_B$. This fact is consistent with the magnetization value when the orbital moment is quenched, and the magnetic moment is approximately given by spin only. The differences between the absolute values of $H_s$ and $M_s$ for the different field directions should be due to the anisotropy of the $g$-factor.

Notable feature is that the magnetization curve for $H \parallel c$ (and $H \parallel b$) has a plateau at approximately 1/3 of $M_s$ between the field $H_{c1}$ and $H_{c2}$ which have been determined from the sharp peaks of $dM/dH$. The plateau is absent for $H \parallel a$. A

![Magnetization and its derivative](https://example.com/magnetization.png)

Fig. 2. (a) Magnetization and its field derivative $dM/dH$ of Cs$_2$CuBr$_4$ for the field parallel to the $c$-axis. (b) The $b^*$-component of the ordering vector $q_0$ as a function of magnetic field $H \parallel c$. Dashed lines denote the lower and higher edge fields of the magnetization plateau. The inset shows $q_0$ around the plateau region.
couple of peaks labeled as $H_{c3}$ and $H_{c4}$ also is suggestive of the tiny magnetization plateau. The level of the tiny plateau is slightly larger than $2/3$ of $M_s$. Since the field range of the second plateau is comparable to the measuring temperature, the plateau is not clearly seen in the magnetization curve. We are planning the experiment which explores the second plateau phase using static magnetic field and dilution refrigerator. The field values of the magnetization plateaux and the saturation fields are summarized in Table I.

Magnetic Bragg reflection appears at $Q = (h, k \pm 0.575, 0)$ \(^{11}\) with integer values of $h$ and $k$ at zero field. This result indicates that the magnetic structure is incommensurate with the ordering vector $q_0 = (0, q_0, 0)$ with $q_0 = 0.575$. The origin of the incommensuration is attributed to the competition between $J_1$ and $J_2$ shown in Fig. 1. Within the classical spin model, the value of $q_0$ is given by $\cos(\pi q_0) = -J_2/(2J_1)$. Using $q_0 = 0.575$, we obtain $J_2/J_1 = 0.467$ for Cs$_2$CuBr$_4$. Figure 2(b) shows the $b^*$-component of the ordering vector $q = (0, q, 0)$ as a function of magnetic field $H \parallel c$. Dashed lines denote the lower and higher edge fields of magnetization plateau at $M \simeq M_s/3$. The value of $q$ increases with increasing magnetic field, and is locked at $q \simeq 2/3$ which is almost commensurate position in the plateau region. This result indicates that the $uud$ spin structure or closely related structure is realized in the plateau phase.

Figure 3 shows the contour plot of the measured $S(Q, \omega)$ mapped on $(k, \omega)$ space. Since the quantum fluctuation is significant in the present system because of $S = 1/2$ and quasi-2D nature, the “effective” value of $\tilde{J}_1$ and $\tilde{J}_2$ which can be obtained from the magnetic excitation at zero field are largely renormalized from the “real” values of $J_1$ and $J_2$. For isostructural Cs$_2$CuCl$_4$, Coldea et al.\(^{9,10}\) determined real exchange constants from the dispersion data in the saturated state. They have obtained $J_1$ and $J_2$ as 0.374(5) meV and 0.128(5) meV, respectively. Since the effective values measured at zero field are $\tilde{J}_1 = 0.61(1)$ meV and $\tilde{J}_2 = 0.107(10)$ meV, the renormalization factor is obtained as $\tilde{J}/J = 1.63(5)$ which

<table>
<thead>
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<th>direction</th>
<th>$H_{c1}$ /T</th>
<th>$H_{c2}$ /T</th>
<th>$H_{c3}$ /T</th>
<th>$H_{c4}$ /T</th>
<th>$H_s$ /T</th>
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<td>–</td>
<td>–</td>
<td>–</td>
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</tr>
<tr>
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<td>28.75</td>
</tr>
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</table>
is comparable to that of \( S = 1/2 \) Heisenberg linear chain. This result supports the existence of strong quantum fluctuation for the present system. The dot-dashed line indicates the spin-wave mode of the helical spin structure\(^{12} \) \( \omega(k) = \sqrt{(J_k - J_{q_0})[(J_k+q_0 + J_k-q_0)/2 - J_{Q_0}]} \) of the Hamiltonian \( \mathcal{H} = J_1 \sum_{i,i'} S_i \cdot S_{i'} + J_2 \sum_{i,j} S_i \cdot S_j \), where \( J_k \) denotes the Fourier transformation of the exchange interaction \( J_k = J_1 \cos(2\pi k) + 2J_2 \cos(\pi k) \cos(\pi \ell) \). In Fig. 3, \( J_2 \) is fixed as \( J_2 = -2J_1 \cos(\pi q_0) \), and \( J_1 \) is scaled as \( J_1 = 2.1 \) meV. Weihong et al.\(^\text{13} \) investigated theoretically the renormalization effect as a function of \( J_2/J_1 \) using the series expansion method. According to their result, the ratio of the real exchange coupling is \( J_2/J_1 = 0.74 \) for \( q_0 = 0.575 \). Since \( J_2/J_1 = 0.37 \) for \( \text{Cs}_2\text{CuCl}_4 \) obtained from the theoretical result by Weihong et al. is in good agreement with \( J_2/J_1 = 0.34 \) (this value is about twice as large as \( J_2/J_1 = 0.175 \) which was derived from the classical spin model with \( q_0 = 0.528 \)). obtained by Coldea et al.\(^9,10 \) the value of \( J_2/J_1 = 0.74 \) should be close to the real value for \( \text{Cs}_2\text{CuBr}_4 \). The dotted line indicates the estimated upper boundary of the continuum scattering generated by the deconfinement of \( S = 1/2 \) spinon pair that is clearly observed for \( \text{Cs}_2\text{CuCl}_4 \).\(^\text{10} \) More decisive experiment which determines the energy extents of the 2-spinon continuum is now ongoing.

§4. Conclusions

The quantum-fluctuation-assisted 1/3 and 2/3 magnetization plateaux were observed on \( \text{Cs}_2\text{CuBr}_4 \) irrespective of the field direction within the bc-plane. The ordering vector was found to be \( q_0 = (0,q_0,0) \) with \( q_0 = 0.575 \) below the ordering temperature \( T \leq 1.4 \) K. In the field parallel to the c-axis, the value of \( q \) is locked at approximately \( q = 2/3 \) in the plateau field region. It can be concluded that the collinear (uud) spin structure is realized in the 1/3 plateau phase. This collinear spin structure is stabilized by the quantum fluctuation. From the inelastic neutron scattering, we have roughly estimated the effective exchange interaction as \( J_1 \approx 2.1 \) meV and \( J_2 \approx 0.98 \) meV.

References