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Magnetization plateaux of the $S = 1/2$ two-dimensional frustrated antiferromagnet Cs_2CuBr_4

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Abstract
The field induced magnetic phase transitions of Cs_2CuBr_4 were investigated by means of a magnetization process and neutron scattering experiments. This system undergoes a magnetic phase transition at Néel temperature $T_N = 1.4$ K at zero field, and exhibits a magnetization plateau at approximately one third of the saturation magnetization for the field directions $H \parallel b$ and $H \parallel c$. In the present study, an additional symptom of the two-third magnetization plateau was found in the field derivative of the magnetization process. The magnetic structure was found to be incommensurate with the ordering vector $\boldsymbol{Q} = (0, 0.575, 0)$ at zero field. With increasing magnetic field parallel to the c-axis, the ordering vector increases continuously and is locked at $\boldsymbol{Q} = (0, 0.662, 0)$ in the plateau field range 13.1 T $< H <$ 14.4 T. This indicates that the collinear *up–up–down* spin structure is stabilized by quantum fluctuation at the magnetization plateau.

1. Introduction

Over the last two decades, the triangular antiferromagnetic (TAF) system has been studied extensively using hexagonal ABX_3-type antiferromagnets, and many types of phase transitions due to the frustration have been found [1]. Within the classical model, the spin structure of the two-dimensional Heisenberg TAF in a magnetic field cannot be determined uniquely, because the number of the parameters that are given by the equilibrium conditions is insufficient for

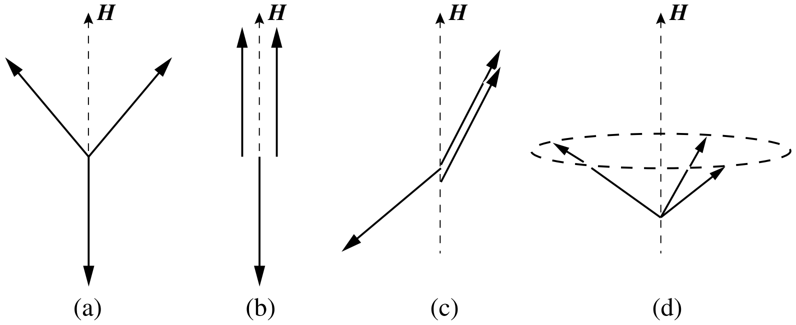

Figure 1. The spin structure of 2D TAF in a magnetic field. (a) Low-field coplanar structure, (b) *up–up–down* structure, (c) high-field coplanar structure and (d) umbrella structure.

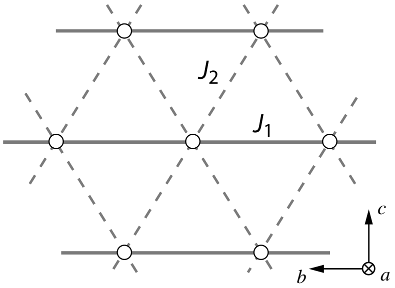

Figure 2. Antiferromagnetic interactions J_1 and J_2 within the bc-plane. The open circles denote Cu^{2+} ions.

the determination of the spin configuration, and thus there remains 'non-trivial continuous degeneracy' at the ground state [2].

Chubukov and Golosov [2] suggested that quantum fluctuation can lift the continuous degeneracy at the ground state. They argued that the Heisenberg TAF undergoes a successive phase transition as shown in figures 1(a)–(c) in that order with increasing magnetic field. The intermediate collinear spin structure is stabilized in a finite field region and the magnetization plateau arises at one-third of the saturation magnetization.

The magnetic phase transition of TAF induced by quantum fluctuation was first found in $CsCuCl_3$ [3, 4]. Due to the weak easy-plane anisotropy, the umbrella spin structure as shown in figure 1(d) is realized in the low-field region for $H \parallel c$-axis, and the spin structures such as (a) and (b) are hidden by the magnetic anisotropy. Consequently $CsCuCl_3$ undergoes a field induced phase transition from the structure (d) to (c), which is accompanied by a small magnetization 'jump' [3].

In Cs_2CuBr_4, the arrangement of $S = 1/2$ Cu^{2+} ions can be regarded as distorted TAF in the bc-plane as shown in figure 2 [5, 6]. The magnetic properties of the isostructural Cs_2CuCl_4 have been investigated extensively by Coldea *et al* [7–10], and Cs_2CuCl_4 was characterized as two-dimensional (2D) TAF. Therefore, it is expected that Cs_2CuBr_4 is also described by an $S = 1/2$ 2D TAF.

In our previous measurements for Cs_2CuBr_4 [5, 6], magnetic ordering was found at $T_N = 1.4$ K, which is more than twice as high as $T_N = 0.62$ K in Cs_2CuCl_4 [7]. The notable feature of the present system is the magnetization plateau at one-third of the saturation magnetization M_S observed for the magnetic field $H \parallel b$ and the c-axes in the ordered phase as shown below. Since the magnetization plateau is observed for two different field directions, its origin is attributed not to the uniaxial magnetic anisotropy but to the quantum effect. On the other hand, no plateau is observed for Cs_2CuCl_4. To the best of our knowledge, the magnetization plateau observed in Cs_2CuBr_4 is the first experimental example of the one-third plateau stabilized by quantum fluctuation in TAF. In the present study, we have remeasured the magnetization process in order to improve the signal-to-noise ratio and performed elastic neutron scattering experiments in order to determine the magnetic structure of Cs_2CuBr_4 in the field parallel to the c-axis up to the plateau region.

2. Experiments

Single crystals of Cs_2CuBr_4 were grown by the slow evaporation method from an aqueous solution of CsBr and $CuBr_2$ in the molar ratio 2:1. The high-field magnetization measurement was performed using an induction method with a multilayer pulse magnet at the Ultra-High Magnetic Field Laboratory, Institute for Solid State Physics, The University of Tokyo. Magnetization data were collected at $T = 400$ mK in magnetic fields up to 35 T.

The elastic neutron scattering experiments at zero field were performed at the HER and LTAS triple axis spectrometer installed at JAERI, Tokai. A single crystal of $\sim$3 cm^3 was used for this measurement.

The field dependence of the elastic neutron scattering experiments were performed at the E1 triple axis spectrometer installed at the experimental reactor of the Hahn-Meitner-Institute, Berlin. The sample used was approximately 1 g in mass. The sample was mounted on the sample stage of the dilution stick DS-X and the dilution stick was loaded into a VM-1 14.5 T vertical superconducting magnet. Measurements were performed for the (a, b) horizontal scattering plane.

3. Results and discussion

3.1. The magnetization process

Figure 3 shows the field dependences of $\mathrm{d}M/\mathrm{d}H$ and the magnetization of Cs_2CuBr_4 for the field direction H parallel to the a-, b- and c-axes measured at $T = 400$ mK. The data were taken in a sweeping down magnetic field. The magnetization saturates at $H_\mathrm{S} \approx 30$ T. The difference between the absolute values of the saturation fields and the saturation magnetizations for the three different field directions should be due to the anisotropy of g-factor. For $H \parallel a$, $\mathrm{d}M/\mathrm{d}H$ does not have any anomaly up to the saturation field. On the other hand, for $H \parallel b$ and $H \parallel c$, $\mathrm{d}M/\mathrm{d}H$ shows several sharp peaks indicated by the arrows $H_{\mathrm{c}1}$–$H_{\mathrm{c}4}$ in figure 3(a). The anomalies $H_{\mathrm{c}1}$ and $H_{\mathrm{c}2}$ correspond to 1/3 magnetization plateau. The level of the 1/3 magnetization plateau is slightly lower than one third of the saturation magnetization $M_\mathrm{S}/3$. A couple of peaks labelled as $H_{\mathrm{c}3}$ and $H_{\mathrm{c}4}$ which are seen for $H \parallel b$ and $H \parallel c$ also have the feature of the magnetization plateau. At $H_{\mathrm{c}3}$ and $H_{\mathrm{c}4}$, the magnetization is just above $2M_\mathrm{S}/3$. For a magnetic system that does not have uniaxial anisotropy, the appearance of a magnetization plateau can be attributed to the existence of a quantum excitation energy gap. Since the distance of the field between these two peaks $H_{\mathrm{c}3}$ and $H_{\mathrm{c}4}$ is approximately 0.6 T, the energy gap around 2/3 plateau $\Delta_{2/3}$ is roughly estimated as $\Delta_{2/3}/k_\mathrm{B} \sim 0.4$ K which is comparable with the measuring temperature. Therefore, the finite temperature effect cannot be neglected for the 2/3-magnetization plateau. In order to confirm the existence of the 2/3-magnetization plateau, measurements at lower temperature are in progress.

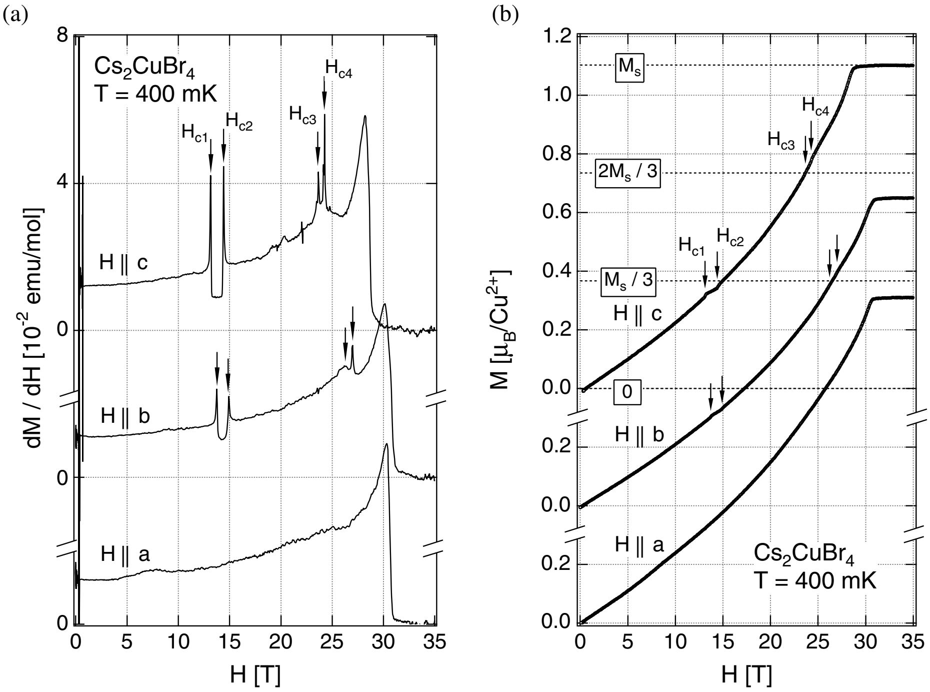

Figure 3. (a) The field derivative of the magnetization and (b) the magnetization as a function of the external field applied for H $\parallel$ the a-, b- and c-axes. Measurements have been done at $T = 400$ mK.

3.2. Neutron scattering

Figure 4(a) shows a typical scan profile along the b^*-direction at around $T = 60$ mK. A magnetic Bragg reflection appears at $\boldsymbol{Q}_0 = (0, 0.575, 0)$. This result indicates that the magnetic structure is incommensurate with the lattice along the b-axis. The origin of the incommensuration is attributed to the competition between J_1 and J_2 (see figure 2). The ordering vector for Cs_2CuCl_4 is $\boldsymbol{Q}_0 = (0, 0.528, 0)$ [7]. In classical theory, the ordering vector $\boldsymbol{Q}_0 = (0, q, 0)$ is given by $\cos(\pi q) = -J_2/(2J_1)$. Using this equation, we obtain $J_2/J_1 = 0.467$ for Cs_2CuBr_4 and $J_2/J_1 = 0.175$ for Cs_2CuCl_4. This result implies that Cs_2CuBr_4 is more frustrated than Cs_2CuCl_4. Of course, since quantum effects cannot be neglected in the present system, we should determine the exchange interactions from the magnetic excitation.

With increasing magnetic field parallel to the c-axis, the value of the ordering vector q increases continuously as shown in figure 4(b). On the other hand, the integrated intensity decreases gradually up to the transition field H_{c1} and shows a jump just after the transition field H_{c1} as shown in figure 4(c). The neutron scattering cross section reflects the spin component perpendicular to the scattering vector. Since the magnetization process for $H \parallel b$ and $H \parallel c$ is almost the same, it can be deduced that the magnetic moments almost lie in the bc-plane as observed in Cs_2CuCl_4 [7]. Thus, the integrated intensity is expected to be increased with increasing external field, because of the increase of the spin component parallel to the c-axis. The reason for the decrease of the intensity for $H < H_{c1}$ is unclear.

Figure 4(b) shows the external field dependence of the ordering vector $\boldsymbol{Q}_0 = (0, q, 0)$ measured for $H \parallel c$-axis. The value of the ordering vector q shows a steep increase just

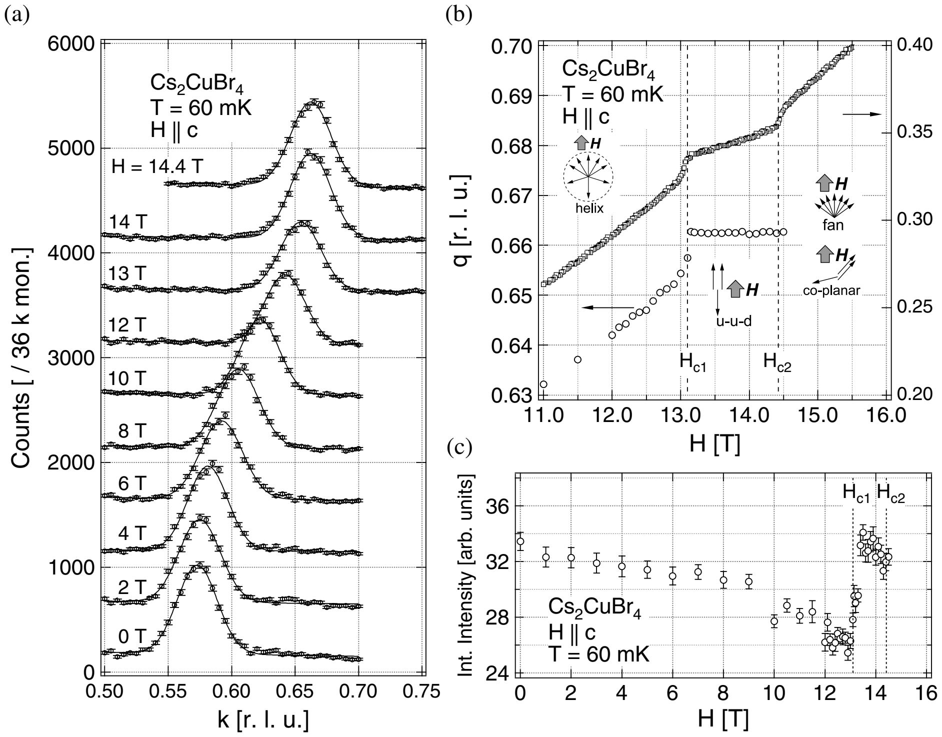

Figure 4. (a) Magnetic Bragg reflections scanned along $\boldsymbol{Q} = (0, k, 0)$ at various external fields. (b) The field dependence of the ordering vector $\boldsymbol{Q} = (0, q, 0)$ (left-hand abscissa) and the magnetization process (right-hand abscissa) around the plateau region. The magnetization curve was measured at $T \sim 400$ mK. (c) The field variation of the integrated intensities of the primary magnetic Bragg peak.

below the lower edge field $H_{c1} = 13.15$ T, and is locked at $q = 0.662$ above $H > H_{c1}$. In figure 4(b), the magnetization curve measured at $T \sim 400$ mK (right-hand abscissa) is also plotted for comparison. The values of H_{c1} determined from the magnetization and the present neutron scattering measurements show excellent agreement. This fact implies obviously that the three-sublattice *up–up–down* spin structure parallel to the external field is realized at the plateau. Measurements have been done up to $H = 14.5$ T; however, any evidence of the magnetic phase transition at $H = H_{c2}$ could not be observed. The transition field H_{c2} seems to be just above the field range of the present measurement.

For the classical helical spin system, a transition from a helical spin structure to a fan structure (see the illustration in figure 4(b)) can occur when an external field is applied in the easy plane [11]. The magnetization process of $RbCuCl_3$ which is described as ferromagnetically stacked distorted TAF shows a *helix–fan* transition for a field parallel to the triangular plane [12, 13]. For $RbCuCl_3$, the quantum fluctuation should be suppressed by the three-dimensional interaction. There is a possibility that the fan spin structure is also realized in Cs_2CuBr_4 for the field region around the saturation field. The fan structure is characterized by the two ordering components parallel and perpendicular to the field. Thus, the Bragg reflections should split from $q = 0.662$, if the fan structure was realized in the high-field region $H > H_{c2}$. If the high-field coplanar structure that has the advantage of quantum fluctuation (shown in figure 1(b)) was realized in the field region $H > H_{c2}$, the ordering vector does not move from $q = 0.662$. In order to confirm the spin structure in the field range $H > H_{c2}$, measurements in higher fields are necessary.

4. Conclusions

We have presented the results of the magnetization process and elastic neutron scattering experiments of Cs_2CuBr_4. It was found that the magnetization curves have plateaux at approximately 1/3 and 2/3 of the saturation magnetization. At zero field, the spin structure in the ordered phase is incommensurate with the ordering vector $\boldsymbol{Q}_0 = (0, q, 0)$ with $q = 0.575$. With increasing field parallel to the c-axis, the value of q is increased, and then locked at approximately $q = 2/3$ above the lower edge field $H_{c1} = 13.15$ T of the magnetization plateau. Thus it can be concluded that the collinear (*up–up–down*) spin structure is realized in the 1/3 plateau field region. This collinear spin structure is stabilized by quantum fluctuation. The spin structures in the higher field region are unclear at present.

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