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Abstract. Cs_2CuBr_4 and $Ba_3NiSb_2O_9$ are magnetically described as quasi-two-dimensional triangular-lattice antiferromagnets with spin-$\frac{1}{2}$ and 1, respectively. We show that both systems exhibit a magnetization plateau at one-third of the saturation magnetization M_s due to the interplay of spin frustration and quantum fluctuation. In Cs_2CuBr_4 that has a spatially anisotropic triangular lattice, successive magnetic-field induced quantum phase transitions including a magnetization plateau $\frac{2}{3}M_s$ were observed. For $Ba_3NiSb_2O_9$, we performed exact diagonalization for rhombic spin clusters with up to 21-sites to analyze the magnetization process. The calculated results are in agreement with experimental observations.

1. Introduction

Triangular-lattice antiferromagnet (TLAF) often exhibits remarkable quantum effect in magnetic field. A typical quantum effect predicted for small spin Heisenberg TLAF is magnetization plateau at one-third of the saturation magnetization M_s [1–3]. For classical spin, the stable state is not uniquely determined, because the number of parameters that determine the spin configuration is more than the number of equations giving the equilibrium conditions. Thus, the ground state is infinitely degenerate. No phase transition accompanied by the anomaly of magnetization arises up to saturation, so the magnetization curve is monotonic, as shown by dashed line in Fig. 1(a). This classical degeneracy can be lifted by the quantum fluctuation that is remarkable for small spin, and a specific spin state is selected as the ground state. Consequently, the *up-up-down* state is stabilized in a finite field range, which leads to a magnetization plateau at $\frac{1}{3}M_s$. Solid line in Fig. 1(a) shows the schematic magnetization curve for the quantum Heisenberg TLAF. The magnetization plateau is a new macroscopic quantum phenomenon.

In contrast to theoretical studies, the experimental studies on the quantum magnetization plateau in TLAF are limited. In the case of spin-$\frac{1}{2}$ and 1, well defined quantum magnetization plateau has not been reported except for Cs_2CuBr_4 and $Ba_3NiSb_2O_9$. In this paper, we show that these two triangular-lattice antiferromagnets exhibit the nonclassical magnetization plateau

at $\frac{1}{3}M_{\rm s}$ [4–7]. In Cs_2CuBr_4, an additional quantum plateau was also observed at $\frac{2}{3}M_{\rm s}$, as shown below [5, 6]. Furthermore, precise thermodynamic measurements revealed cascade of field-induced quantum phase transitions [8, 9].

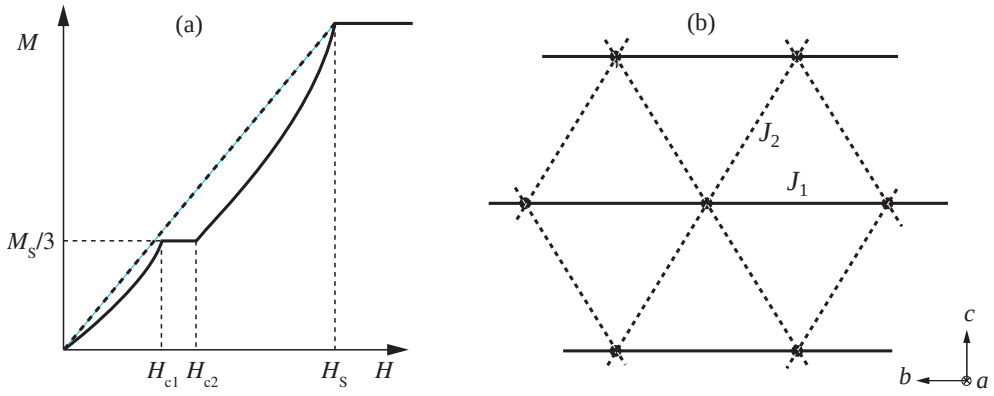

Figure 1. (a) Schematic magnetization curves of triangular-lattice Heisenberg antiferromagnets with classical (dashed line) and quantum spins (solid line). (b) Spatially anisotropic triangular lattice composed of Cu^{2+} ions in Cs_2CuBr_4.

2. Spatially anisotropic S=1/2 triangular-lattice antiferromagnet Cs_2CuBr_4

Cs_2CuBr_4 has an orthorhombic structure with space group $Pnma$ [10, 11]. The structure is composed of $CuBr_4^{2-}$ tetrahedra and Cs^+ ions. Magnetic Cu^{2+} ions that have spin-$\frac{1}{2}$ form a distorted triangular lattice parallel to the bc plane. Figure 1(b) shows the exchange network in the bc plane. There are two kinds of antiferromagnetic nearest-neighbor exchange interaction J_1 and J_2.

Cs_2CuBr_4 undergoes three-dimensional magnetic ordering at $T_{\rm N} = 1.4\,{\rm K}$ [4]. In the ordered phase, spins lie approximately in the bc plane and form an incommensurate helical structure that is characterized by a wave vector $\boldsymbol{Q}_0 = (0, 0.575, 0)$. 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_2CuBr_4. Because of spin-$\frac{1}{2}$, the quantum correction of the helical pitch is significant in the present system, and thus, the value of J_2/J_1 obtained from the classical spin model is different from the real value when $J_2 \neq J_1$. Zheng *et al.* [12] investigated theoretically a spatially anisotropic Heisenberg TLAF with spin-$\frac{1}{2}$, which is equivalent to the model shown in Fig. 1(b). They calculated the ordering vector Q_0 as a function of $J_1/(J_1 + J_2)$. From their result and $Q_0 = 0.575$, we obtain $J_2/J_1 = 0.74$, which is 1.6 times as large as the classical value.

Figure 2(a) shows the magnetization curve and dM/dH vs H measured at $T = 0.4$ K for $H \parallel c$. Notable feature is that the magnetization curve has a plateau at approximately one-third of the saturation magnetization $M_{\rm s}$. This is more clearly recognized in dM/dH vs H shown in Fig. 2(a). Derivative susceptibility dM/dH exhibits sharp peaks at the plateau edges. The lower and higher edge fields are $H_{c1} = 13.1$ and $H_{c2} = 14.4\,{\rm T}$, respectively. For $H_{c1} < H < H_{c2}$, dM/dH is small. The $\frac{1}{3}$-magnetization-plateau was also observed for $H \parallel b$, while no plateau was observed for $H \parallel a$. Because the $\frac{1}{3}$-magnetization-plateau in Cs_2CuBr_4 is clearly observed for two different field directions, the plateau cannot be explained in terms of the classical model. Thus, we can conclude that the magnetization plateau arises from the quantum effect. The Dzyaloshinsky-Moriya (DM) interaction with the $\boldsymbol{D}$ vector perpendicular to the triangular lattice should be responsible for the absence of the magnetization plateau for $H \parallel a$.

The magnetic-field dependence of the ordering vector Q_0 was measured through neutron elastic scattering [5, 6]. The value of Q_0 increases with increasing magnetic field, and is locked

at $Q_0 \simeq 2/3$ in the plateau region. This result indicates that the *up-up-down* spin structure is realized in the plateau state.

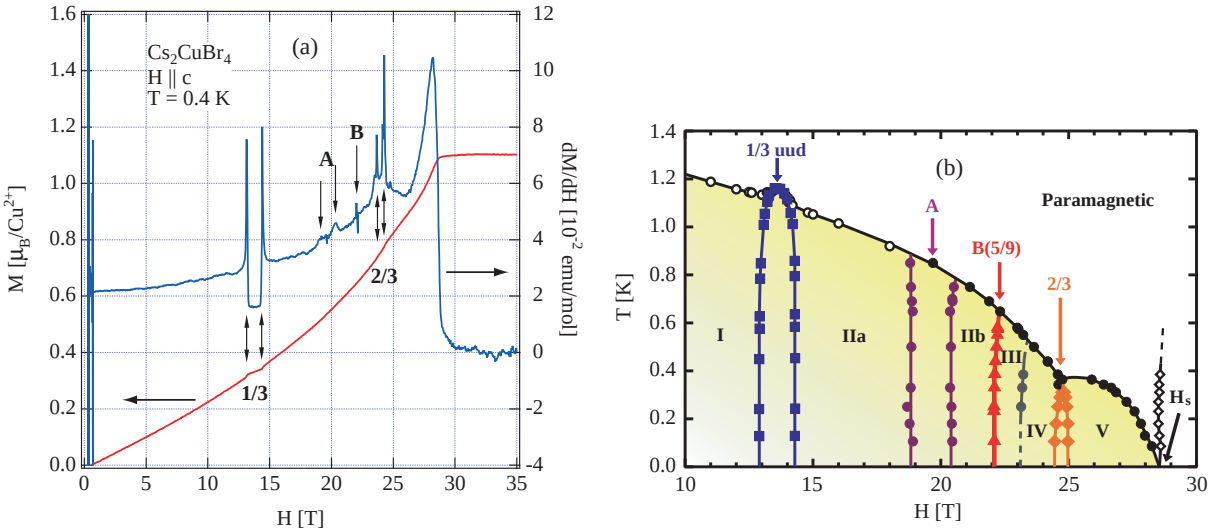

Figure 2. (a) Magnetization curve and dM/dH vs H in Cs_2CuBr_4 measured at $T = 0.4\,\mathrm{K}$ for $H \parallel c$. Arrows denote critical fields. (b) Magnetic phase diagram obtained from the magnetocaloric-effect data measured at various temperatures for $H \parallel c$.

Fujii *et al.* [13, 14] performed ^{133}Cs-NMR experiments on Cs_2CuBr_4 in the magnetic fields that cover the field range of the 1/3 magnetization plateau. For $H \parallel b$, they observed that outside the plateau region, the NMR spectrum has a double-horn type broad shape characteristic of an incommensurate spin structure, while in the plateau region, the spectrum changes into discrete two peaks indicative of a commensurate *up-up-down* spin structure. Their NMR results are consistent with magnetization and neutron scattering measurements.

As shown in Fig. 2(a), dM/dH exhibits additional sharp double peak structure around $H \sim 23$ T, where the magnetization is approximately $\frac{2}{3}M_s$. This is indicative of the second tiny plateau at $\frac{2}{3}M_s$. This additional sharp double peak structure in dM/dH was also observed for $H \parallel b$. The magnetization processes for $H \parallel c$ and b almost coincide when normalized by the g factor. This means that spin-spin interactions are almost isotropic in the triangular-lattice plane.

Miyahara *et al.* [15] investigated the ground state of the spatially anisotropic Heisenberg TLAF with spin-$\frac{1}{2}$ by exact diagonalization. They showed that the $\frac{1}{3}$- and $\frac{2}{3}$-magnetization-plateaus appear for $0.7 \leq J_2/J_1 \leq 1.3$ and $0.5 \leq J_2/J_1 \leq 0.8$, respectively. Fortunately, the value of $J_2/J_1 = 0.74$ for Cs_2CuBr_4 satisfies both conditions. This should be the reason why both $\frac{1}{3}$- and $\frac{2}{3}$-magnetization-plateaus were observed in Cs_2CuBr_4.

Fortune *et al.* [9] performed magnetocaloric effect and magnetic-torque measurements and found the cascade of magnetic-field induced quantum phase transitions. Figure 2(b) is the phase diagram for $H \parallel c$ determined by magnetocaloric effect. In addition to the $\frac{1}{3}$- and $\frac{2}{3}$-plateau phases, many quantum phases were observed. The phase transitions to the A phase and the transitions to the very narrow B phase can also be observed as small peaks in dM/dH shown in Fig. 2(a). These successive quantum phase transitions should be attributed to the spatially anisotropic triangular lattice and the Dzyaloshinsky-Moriya interaction with the $\boldsymbol{D}$ vector perpendicular to the triangular lattice [15–19], but the overall explanation is still an open question.

3. S=1 triangular-lattice antiferromagnet $Ba_3NiSb_2O_9$

Figure 3 shows the crystal structure of $Ba_3NiSb_2O_9$. This compound crystalizes in a high symmetric hexagonal structure, $P6_3/mmc$ [20, 21], which is the same as the hexagonal $BaTiO_3$ structure. The structure is composed of single NiO_6 octahedra and face-connected Sb_2O_9 double octahedra, which are linked sharing corners. Magnetic Ni^{2+} ions that have spin-1 form triangular lattice parallel to the c plane.

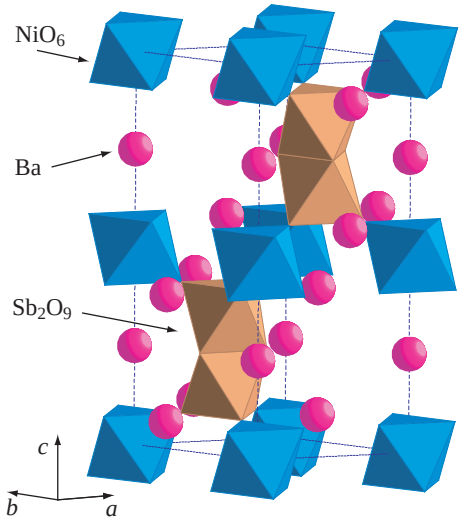

Figure 3. Crystal structure of $Ba_3NiSb_2O_9$. Dotted lines denote the chemical unit cell.

The magnetic properties of $Ba_3NiSb_2O_9$ were investigated by Doi *et al.* [22]. They reported that $Ba_3NiSb_2O_9$ undergoes antiferromagnetic ordering at $T_N = 13.5\,K$. In the present study, we first measured specific heat to investigate the nature of the magnetic ordering. We found that $Ba_3NiSb_2O_9$ undergoes two magnetic phase transitions at $T_{N1} = 13.5\,K$ and $T_{N2} = 13.0\,K$. In Heisenberg-like TLAF, the successive phase transition arises when the magnetic anisotropy is of easy-axis type, while a single transition occurs for the easy-plane anisotropy [23]. The successive phase transitions with a very narrow intermediate phase in $Ba_3NiSb_2O_9$ indicates that the magnetic anisotropy is of the easy-axis type and is much smaller than the exchange interaction. The origin of the anisotropy should be single ion anisotropy of the form $D(S^z)^2$.

Figure 4(a) shows the magnetization curve of $Ba_3NiSb_2O_9$ powder and dM/dH vs magnetic field H measured at 1.3 K. A well defined magnetization plateau is observed at $M \simeq 0.8\,\mu_B/Ni^{2+}$. Because the g factor of $Ba_3NiSb_2O_9$ powder is $g \simeq 2.3$ [24], the magnetization at the plateau corresponds to one-third of the saturation magnetization M_s. The magnetization anomalies at the lower and the higher edge fields are smeared because of powder sample.

In the case of classical Heisenberg TLAF with the easy-axis anisotropy along the c axis, the $\frac{1}{3}$-plateau appears for $H \parallel c$, while not for $H \perp c$ [25]. Thus, for powder sample of such classical system, dM/dH at the plateau range should be about two-third of dM/dH for slope region below the lower edge field. In the present system, the minimum of dM/dH at the plateau range is smaller than half of dM/dH for $H < 30\,T$. This implies that the $\frac{1}{3}$-plateau appears, irrespective of external field direction. In classical Heisenberg TLAF, thermal fluctuation stabilizes a collinear *up-up-down* state at finite temperatures, so that plateau-like magnetization anomaly occurs in magnetization curve [26]. The magnetic field range of the thermally stabilized collinear state decreases with decreasing temperature and becomes zero at $T = 0$. The $\frac{1}{3}$-plateau in $Ba_3NiSb_2O_9$ has a magnetic field range of about 10 T even at 1.3 K ($\ll T_{N2}$). Thus, the $\frac{1}{3}$-plateau observed in $Ba_3NiSb_2O_9$ arises from the quantum effect. However, the magnetization at the plateau range

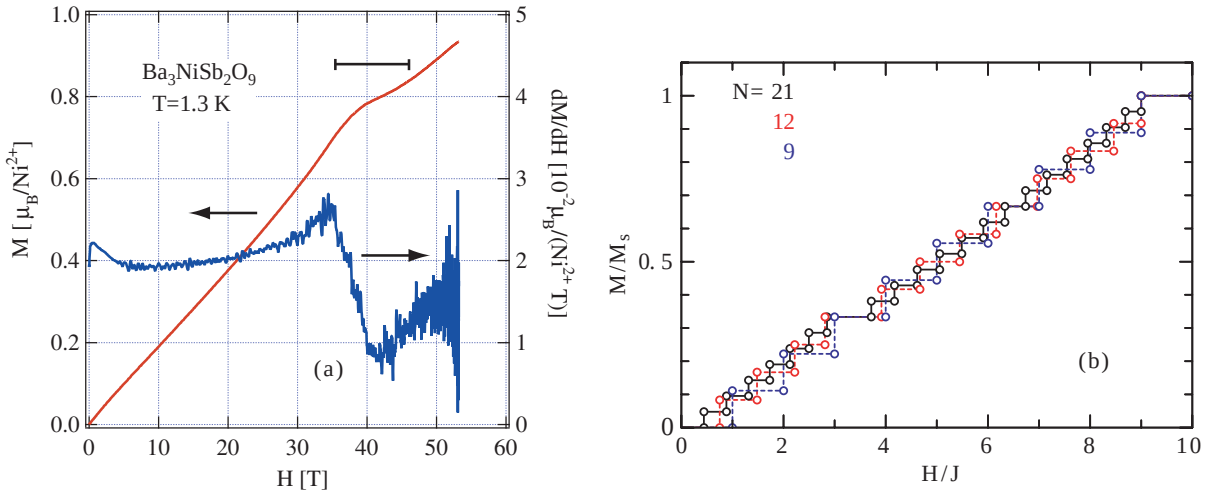

Figure 4. (a) Magnetization curve and dM/dH vs H of $Ba_3NiSb_2O_9$ measured at 1.3 K. A horizontal bar denotes the field range of plateau calculated by exact diagonalization. (b) Magnetization curves for spin-1 Heisenberg TLAF calculated by exact diagonalizations for 9-, 12-, and 21-site rhombic clusters.

is not completely flat. This should be ascribed to the increase of the sample temperature due to the magnetocaloric effect, as observed in Cs_2CuBr_4.

Figure 4(b) shows magnetization curves for spin-1 Heisenberg TLAF calculated by exact diagonalization for 9-, 12- and 21-site rhombic clusters. The calculated magnetization curve shows a plateau at $\frac{1}{3}M_s$, although the magnetic field range of the plateau is fairly small as compared with the spin-$\frac{1}{2}$ case [2, 3]. The lower and higher edge fields tend to converge to $H_{c1} = 2.85J$ and $H_{c2} = 3.72J$, respectively, with increasing cluster size. A horizontal bar in Fig. 4(a) denotes the calculated field range of the $\frac{1}{3}$-plateau. Here we chose the center field of the $\frac{1}{3}$-plateau as $(H_{c1} + H_{c2})/2 = 41$ T. The experimental field range of the plateau is roughly estimated as 70% of the calculated field range. The field range of the $\frac{1}{3}$-plateau observed in $Ba_3NiSb_2O_9$ is consistent with that calculated for spin-1 Heisenberg TLAF.

Läuchli *et al.* [27] also calculated the magnetization curve for spin-1 Heisenberg TLAF using exact diagonalization up to 27-site cluster and showed a small magnetization plateau at $\frac{1}{3}M_s$. However, their calculations for $N \geq 21$ are limited to a certain magnetization range and do not cover all the magnetization value. For spin-1 case, the field range of the $\frac{1}{3}$-magnetization-plateau normalized by the saturation field H_s is calculated as $(H_{c2} - H_{c1})/H_s = 0.097$, while for spin-$\frac{1}{2}$ case, the normalized plateau range is 0.171 [3, 29]. The $\frac{1}{3}$-magnetization-plateau for spin-1 case is considerably suppressed as compared with the spin-$\frac{1}{2}$ case.

For TLAF with $S \geq 1$, the magnetization plateau at $\frac{1}{3}M_s$ can be stabilized with the help of the biquadratic exchange interaction [27, 28], which mainly arises from the spin-lattice coupling as discussed by Penc *et al.* [28]. The biquadratic exchange interaction may be responsible for the magnetization plateau observed in $RbFe(MoO_4)_2$ [30], because the normalized plateau range is as large as 0.10 despite spin-$\frac{5}{2}$. In $Ba_3NiSb_2O_9$, the contribution of the biquadratic exchange interaction should be negligible, because the field range of the magnetization plateau can be explained quantitatively within the the bilinear exchange interaction.

4. Conclusion

We have presented experimental evidence of magnetic-field induced quantum phase transitions in Cs_2CuBr_4, which is described as a spatially anisotropic spin-$\frac{1}{2}$ TLAF. Cs_2CuBr_4 exhibits quantum magnetization plateaus at $\frac{1}{3}M_s$ and $\frac{2}{3}M_s$ for magnetic field parallel to the *bc* plane. In addition to these magnetization-plateau phases, many quantum phases induced by magnetic field were observed as shown in Fig. 2(b). We have also presented the results of high-field magnetization measurements on $Ba_3NiSb_2O_9$ that is described as a spin-1 Heisenberg TLAF. We observed a well defined nonclassical magnetization plateau at $\frac{1}{3}M_s$. We performed exact diagonalization for rhombic spin clusters with up to 21-sites to analyze the magnetization process. The calculated results are in agreement with experimental observations.

Acknowledgments

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