Magnetic field effect on the magnetic torque and the magnetostriction in \((\text{CH}_3)_2\text{CHNH}_3\text{CuCl}_3\)

T Suzuki\(^1\), T Saito\(^2\), T Sasaki\(^3\), A Oosawa\(^2\), T Goto\(^2\), S Awaji\(^3\), K Watanabe\(^3\), N Kobayashi\(^2\) and H Manaka\(^4\)

\(^1\)Advanced Meson Science Laboratory, RIKEN, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan
\(^2\)Department of Physics, Sophia University, 7-1 Kioi-cho, Chiyoda-ku Tokyo, 102-8554, Japan
\(^3\)Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
\(^4\)Graduate School of Science and Engineering, Kagoshima University, Korimoto, Kagoshima 890-0065, Japan

E-mail: suzuki_takao@riken.jp

Abstract. Magnetic torque \(\tau\) and magnetostriction \(\Delta L/L\) measurements were carried out in the spin gap system \((\text{CH}_3)_2\text{CHNH}_3\text{CuCl}_3\). The magnetic torque shows a cusp-like anomaly at the magnetic field \(H_{a1}\) with increasing the magnetic field, and the magnetostriction shows a kink at almost the same magnetic field. These results suggest that anomalous change in the magnetic torque is associated with the lattice distortion, and it is expected that the field-induced magnetic phase transition is not simple Bose-Einstein condensation in this system.

The field-induced magnetic ordering in quantum spin systems has been one of the most attractive issues in the condensed matter physics because the quantum phase transition between the nonmagnetic disordered phase with spin gap and the magnetically ordered phase can be interpreted as the Bose-Einstein Condensation (BEC) of magnons \([1, 2]\).

The title compound \((\text{CH}_3)_2\text{CHNH}_3\text{CuCl}_3\) (abbreviated as IPACuCl\(_3\)) is the spin gap system\([3-7]\). Quite recently, it was reported that this system is characterized as the spin ladder along the \(a\)-axis with the strongly coupled ferromagnetic rungs and that the excitation gap is re-estimated as 13.6 K by means of the neutron inelastic scattering measurements \([8]\).

When a magnetic field is applied in the spin gap system, the Zeeman splitting occurs and then an energy gap between the singlet level and one of the triplet levels vanishes at the field \(H_g\) corresponding to the spin excitation gap. For \(H > H_g\), the ground state becomes magnetic due to the merging of the triplet level into the singlet level, and the system can undergo the magnetic ordering with the help of the three dimensional interactions. Considering the triplet as a magnon which is a boson with \(S = 1\) in the singlet sea treated as a vacuum, the spin gap system can be mapped on the magnon system and the field-induced magnetic ordering can be described as the BEC of magnons \([1, 2]\).

However, the magnon BEC picture is only applied to spin gap systems in which there is rotationally invariance for the direction of the applied magnetic field in order to conserve the number of magnons. This restriction means that the existence of the anisotropy is important to describe the property of the field-induced magnetic ordering, namely whether or not the ordering can be interpreted as the BEC of magnons. In this study, we carried out the magnetic...
torque and the magnetostriction measurements in order to investigate the effect of the magnetic anisotropy on the field-induced magnetic phase transition in IPACuCl$_3$.

The single crystal of IPACuCl$_3$ were grown from dissolving $(\text{CH}_3)_2\text{CHNH}_2\cdot\text{HCl}$ and CuCl$_2$ in isopropyl alcohol by the evaporation method [3]. The crystals with three orthogonal surfaces were obtained. We define the A-, B-, and C-plane as shown in ref. 3. The magnetic torque was measured using a cantilever beam torque meter [9]. The amplitude of the torque is obtained by measuring the capacitance with the use of a lock-in amplifier and a capacitance bridge. The measurements were carried out using the 28 T-HM hybrid magnet with $^3$He refrigerator at High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University. The magnetostriction was measured by the strain gauge method using the relation $\kappa \Delta L/L = \Delta R/R$ ($\kappa = 2.1$, $L = 2$ mm, $R = 118$ $\Omega$). The gauge was glued onto a surface of the single crystal by the apiezon N grease. The field dependence measurements were carried out using the 12 T superconducting magnet settled in Department of Physics, Sophia University.

Figure 1 shows the field dependence of the magnetic torque $\tau$ at $T = 4.0$ K for $H \perp$ C-plane of IPACuCl$_3$ as a typical case. The magnetic torque increases rapidly with increasing magnetic field, and a cusp-like change appears at $H_{a1}$. Above $H_{a1}$, the magnetic torque shows some cusp-like anomalies and then turns to decrease at $H_{a4}$. Figure 2 displays the representative profiles of the field dependence of the magnetic torque at various temperatures below $H = 12$ T under the configuration of $H \perp$ C-plane in IPACuCl$_3$. The cusp-like anomalies at $H_{a1}$ and $H_{a2}$ can be seen clearly at all the temperatures studied in this work, and the large hysteresis is observed between $H_{a1}$ and $H_{a2}$. The magnitude of the magnetic torque $\tau$ at $H_{a1}$ and $H_{a2}$ is almost the same at each temperature. The absolute value of the applied magnetic field at the cusp-like anomaly at $H_{a1}$ is lower than the critical magnetic field determined by the magnetization and the specific heat measurements [4, 5]. This means that the anomaly at $H_{a1}$ appears before the closing of the spin gap, therefore the cusp-like anomaly at $H_{a1}$ seems to be related to the total magnetization, which is the sum of the thermally-exited and the field-induced magnetic moment.

Figure 3 shows the magnetostriction $\Delta L/L$ perpendicular to the B- and A-plane as a function of the applied external magnetic field of $H \perp$ C-plane at $T = 4.0$ K. As shown in Fig. 3, $\Delta L/L$ perpendicular to the B-plane increases, and $\Delta L/L$ perpendicular to the A-plane decreases with increasing the magnetic field. Especially in case of $\Delta L/L$ perpendicular to the B-plane, a kink
is observed at $H = 7.5$ T. This value of the magnetic field $H = 7.5$ T is quite close to $H_{a1}$ where the cusp-like anomaly in the magnetic torque appears, although no anomaly is seen in $\Delta L/L$ at $H_{a2}$ within the resolution. The changed value of $|\Delta L/L|$ in the direction perpendicular to the A-plane is four times as large as the case in the direction perpendicular to the B-plane. Figure 4 shows the magnetostriction $\Delta L/L$ perpendicular to the B-plane at various temperatures. Below $T = 7$ K, $\Delta L/L$ is almost zero in the lower magnetic field region and then begins to increase at $H_L$. The kink point $H_L$ decreases with increasing temperature, and the anomaly vanishes at 10 K.

The characteristic magnetic field $H_{a1}$ determined in the magnetic torque curve and $H_L$ determined in the magnetostriction curve are plotted on the $H$-$T$ plane in Fig. 5. $H_L$ is almost the same with $H_{a1}$ below 4 K, and a plot of $H_L$ at 7 K is on the extrapolated line from the lower temperature side. This result indicates that the anomalies observed in the magnetic torque at $H_{a1}$ and observed in the magnetostriction at $H_L$ have the same origin. As mentioned above, the cusp-like anomaly at $H_{a1}$ and the kink at $H_L$ are observed in the outside of the magnetically ordered region on the phase diagram deduced from the magnetization and the specific heat measurements [4, 5]. Thus, these anomalous changes observed in the magnetic torque and in the magnetostriction are not the phenomena which are directly associated with the field-induced magnetic phase transition. The separation of the change of the magnetostriction and the field-induced magnetic ordering has not been reported in other spin gap systems[10-13].

Figure 6 shows the schematic illustration of the crystal structure and the direction of a lattice distortion. Considering the spacial relation between the crystal grown surfaces (A-, B- and C-plane) and the crystal structure [14], the crystal lattice seems to expand to the direction of $a$-axis, and this distortion corresponds to the increase of the length between rungs of the ladder.

The anomalous change in the magnetostriction and change in the magnetic torque at $H_{a1}$ occur simultaneously. In addition, the hysteresis is observed in the magnetic torque above $H_{a1}$ although no hysteresis is detected in the magnetostriction within the resolution. These things mean that the change of the magnetic property is associated with the crystal distortion, so that the cusp-like change of the magnetic torque is originated from not only a pure change in the
spin system but a change in the crystal lattice system. Thus, because the cross term interaction between the spin system and the crystal lattice system should exist in the Hamiltonian describing the spin state of IPACuCl$_3$, it is expected that the field-induced magnetic phase transition at the higher magnetic field is not simple Bose-Einstein condensation.

In summary, we carried out the magnetic torque and the magnetostriction measurements in the spin gap system (CH$_3$)$_2$CHNH$_3$CuCl$_3$. The magnetic torque shows a cusp-like anomaly at almost the same magnetic field at which a kink appears in the magnetostriction curve with increasing magnetic field. This result suggests that the anomalous change in the magnetic torque is associated with the crystal lattice distortion.

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