High field magnetism of the triangular lattice antiferromagnet $CsFeCl_3$ under high pressure

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Abstract

Magnetization measurements of the triangular lattice antiferromagnet $CsFeCl_3$ at 1.4 K for $H \parallel c$ were performed in pulsed high magnetic fields of up to 51 T under high pressures of up to 0.80 GPa. At ambient pressure, small magnetization below $H_{c1} = 4$ T changes steeply between H_{c1} and H_{c2} =12 T, then showing the magnetization with small slope up to H_m =33 T where metamagnetic transition with two steps occurs. With increasing pressure, the H_{c1} and H_m shifted to the low-field side, while H_{c2} shifted to the opposite side. The change in the H_m may depend on the difference in the single-ion anisotropies on pressure in the lowest $J = 1$ and the excited $J = 2$ states.

1 Introduction

Gapped spin systems with a singlet ground state are attractive quantum spin systems, because the gap is suppressed by the applied magnetic field, pressure, or chemical doping, causing a quantum phase transition (QPT). In the vicinity of the quantum critical point (QCP) where the QPT occurs at zero temperature, some exotic phenomena are expected to be observed by the effect of quantum fluctuations.

$CsFeCl_3$ is one of the hexagonal ABX_3-type families that possesses the magnetic frustration in the triangular-lattice antiferromagnetic plane and belongs to a space group $P6_3/mmc$ [1]. Figure 1(a) shows the crystal structure of $CsFeCl_3$. Magnetic Fe^{2+} ions, which are surrounded octahedrally by six Cl^- ions, form a chain structure with ferromagnetic (FM) exchange interactions (J_0) along the c axis (chain direction) and a triangular lattice with antiferromagnetic (AFM) exchange interactions (J_1) in the ab plane [2]. The energy scheme of Fe^{2+} ion in $CsFeCl_3$ is depicted in Fig. 1(b). The $^5T_{2g}$ lowest state in a cubic crystalline field splits into three states owing to spin-orbit coupling. The energy gap between the lowest $J = 1$ state and the next excited $J = 2$ state is $2\lambda(\approx 200\ \mathrm{cm}^{-1})$ where λ is the spin-orbit coupling constant of Fe^{2+} ion. The $J = 1$ state consists of a

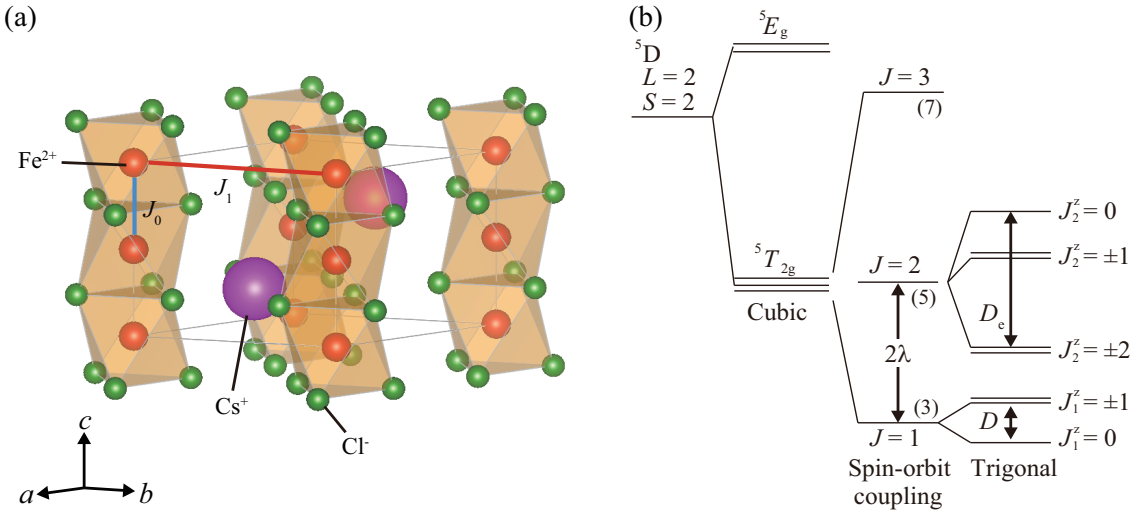

Figure 1: (a) Crystal structure of $CsFeCl_3$. Magnetic Fe^{2+} ions are surrounded octahedrally by six Cl^- ions. $FeCl_6$ octahedra form the chain along the c axis by sharing Cl-triangular faces. The exchange interactions between Fe^{2+} ions are denoted by the intra-chain ferromagnetic constant J_0 and the inter-chain antiferromagnetic constant J_1. (b) Expected energy scheme of Fe^{2+} ion in $CsFeCl_3$

singlet ground state and doublet excited state with a spin gap (Δ_s) owing to easy-plane single-ion anisotropy. Considering only the energy scheme of the $J = 1$ state, the magnetic interaction terms of Fe^{2+} ions (fictitious spin $S = 1$) in $CsFeCl_3$ in magnetic fields applied along the z axis (c direction) are described by the following Hamiltonian [3]

$$\mathcal{H} = \sum_i D(S_i^z)^2 + J_0 \sum_{<i,j>}^{\text{chain}} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + J_1 \sum_{<i,k>}^{\text{plane}} \boldsymbol{S}_i \cdot \boldsymbol{S}_k - g\mu_B H \sum_i S_i^z, \tag{1}$$

where D (>0) is the single-ion anisotropy constant, J_0 and J_1 are the ferromagnetic (along the chain) and antiferromagnetic (in the ab plane) exchange constants, respectively. g and μ_B in the forth term are the g factor for the z axis and the Bohr magneton, respectively. The coupling constants determined from the energy-dispersion relations in the inelastic neutron scattering (INS) experiments [4] are D = 2.18 meV, J_0 = -0.454 meV, and J_1 = 0.024 meV . As aforementioned, $CsFeCl_3$ possesses a singlet ground state with a spin gap to the exicited doublet state owing to this positive D.

With increasing applied pressure at zero field, Δ_s was reported to diminish and then vanish above P_c = 0.90 GPa to exhibit a magnetically ordered state with a 120° spin structure in the ab plane at low temperatures [5, 6]. In the INS experiments of this compound under high pressure, the excitation modes, in which the longitudinal and transverse fluctuations of spins are hybridized, were observed at 1.4 GPa [7].

As for the magnetic-field experiments, the magnetization curve at 1.3 K for $H \parallel c$ increases gradually up to H_{c1} = 4 T, and then increases rapidly between H_{c1} and H_{c2} = 12 T [8]. Neutron scattering experiments in magnetic fields revealed that the ground-state spin configuration is a 120° spin structure in the triangular-lattice plane between H_{c1} and H_{c2} [9]. Above H_{c2}, the magnetization, increases slightly with magnetic field, which is attributed to the Van Vleck paramagnetism, and then exhibits a metamagnetic transition around H_m = 33 T [8]. This metamagnetic transition was suggested to be caused by an unconventional level crossing between the $J_1^z = -1$ and $J_2^z = -2$ states in magnetic fields as indicated in Fig. 1 (b). In this study, we performed high-field magne-

tization measurements of $CsFeCl_3$ at 1.4 K for $H \parallel c$ under pressures of up to 0.80 GPa and constructed the magnetic-field (H) versus pressure (P) phase diagram to discuss the pressure dependence of H_{c1}, H_{c2} and H_m.

2 Experiment

Single crystals of $CsFeCl_3$ were grown by the vertical Bridgman method described in Ref. [5]. The sample was a cylindrical single crystal with 4 mm in length along the c axis and 1.6 mm in diameter in the ab plane. Pulsed high magnetic fields of up to 51 T with the duration of 35 ms were generated with a non-destructive pulsed magnet at AHMF at Osaka University. High-field magnetization measurements were performed with an induction method using a pick-up coil at the temperature of 1.4 K under high pressures of up to 0.80 GPa. In this study, we used a NiCrAl piston-cylinder-type pressure cell (PCC) with inner and outer diameters of 2.0 mm and 6.0 mm, respectively, and a mixture of Fluorinert FC70:FC77 = 1:1 as the pressure medium. The applied pressure in the sample space was calibrated by the pressure dependence of the superconducting transition temperature of Sn [10]. When putting the sample into the PCC, the sample could be adjusted by hand to apply the magnetic field to the c axis of the sample within $\sim 4°$. The magnetization measurement of $CsFeCl_3$ at ambient pressure was performed without the NiCrAl PCC. Pulsed magnetic field induced eddy current in metallic parts of the NiCrAl PCC, resulting in the Joule heating. When the NiCrAl PCC is placed in liquid ^{4}He cryostat at 1.4 K, the temperature in the sample space does not change much until 6.5 ms (approximately 40 T in the ascending process when the maximum field was 51 T) from the start of magnetic-field generation [11], thus allowing high-field measurements up to ~40 T at the lowest temperature in the field ascending process.

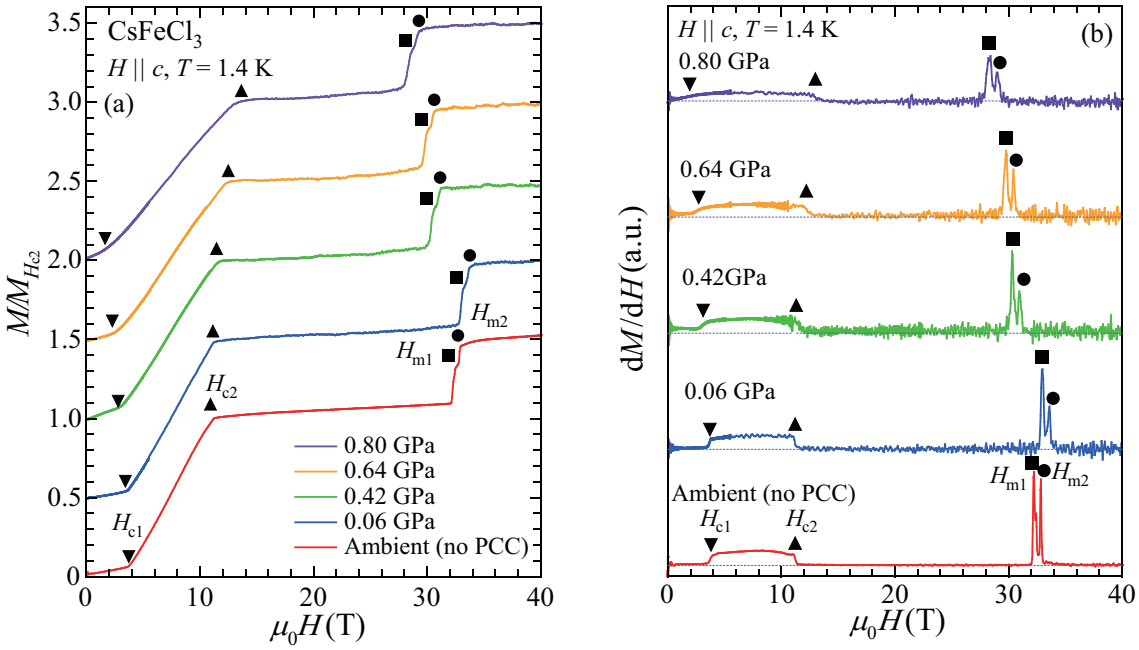

Figure 2: (a) Magnetization (M) curves of $CsFeCl_3$ at 1.4 K for $H \parallel c$ normalized by the M at H_{c2} under various pressures. (b) The field derivatives of the magnetization (dM/dH) curves in (a). Both $M/M_{H_{c2}}$ in (a) and dM/dH in (b) are shifted arbitrarily along the vertical direction for clarity.

3 Results and Discussion

The magnetization (M) curves normalized by the M at H_{c2} and dM/dH of $CsFeCl_3$ at 1.4 K for $H \parallel c$ under various pressures are shown in Figs. 2 (a) and (b), respectively. $M/M_{H_{c2}}$ and dM/dH at ambent pressure are in good agreement with those in a previous paper [8]. The $M/M_{H_{c2}}$ around 33 T exhibits a metamagnetic transition with two steps at $H_{m1} = 32.2$ T and $H_{m2} = 32.9$ T, which corresponds to two peaks in dM/dH. With increasing pressure, the transitions fields H_{c1}, H_{m1} and H_{m2} shift to the low-field side, while the H_{c2} shifts to the high-field side. The change in M (ΔM) or the slope between H_{c2} and the magnetic field just below H_{m1} was close to each other independent of pressure. Therefore, the Van Vleck paramagnetism is almost independent of pressure, suggesting weak pressure dependence of the spin-orbit coupling constant λ.

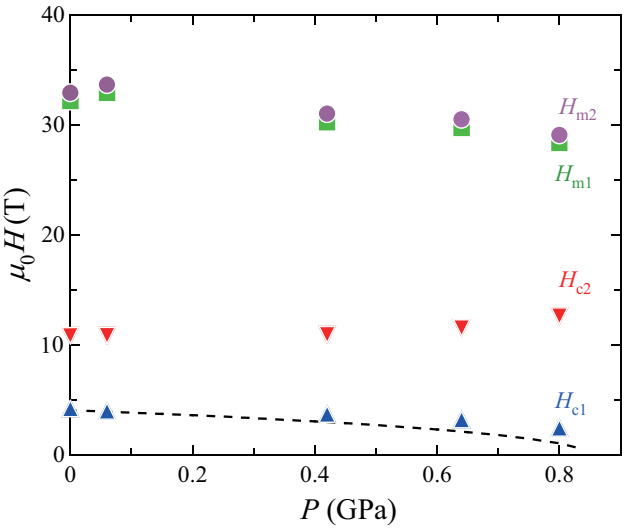

Figure 3: Magnetic field ($\mu_0 H$) vs pressure (P) phase diagram of $CsFeCl_3$ at 1.4 K for $H \parallel c$ constructed from the high-field magnetization measurements. Dashed line is the pressure dependence of H_{c1} provided in a previous study [5].

Figure 3 shows the magnetic field versus pressure phase diagram of $CsFeCl_3$ at 1.4 K for $H \parallel c$. The calculated formula for H_{c1} [5] is given by,

$$H_{c1} = \Delta_s/g\mu_B = \sqrt{D^2 - 2D(2J_0 + 3J_1)}/g\mu_B. \quad (2)$$

The pressure dependences of the exchange interaction and the single-ion anisotropy determined from the INS study [7] were described as follows; J_0 (meV) = -0.5 -0.14P, J_1 (meV) = 0.0312 -0.0015P, and D (meV) = 2.345 +0.365P, where the P (GPa) is the value of pressure. The dashed line in Fig. 3 expresses the pressure dependence of H_{c1} given by Eq. (2), in which the pressure-dependent exchange constants and the single ion anisotropy constant given above are substituted. The comparison of the experimental data to the calculated one is made and these are closely placed to each other. The spin states of Fe^{2+} ion with the fictitious spin $S = 1$ in $CsFeCl_3$ are composed of a singlet-ground state ($S^z = 0$) and the doublet-excited state ($S^z = \pm 1$) separated by a single-ion anisotropy D as shown in Fig. 1(b). However, the excited state has a finite energy band that becomes broad with increasing pressure. Between H_{c1} and H_{c2}, the lower doublet state with a finite bandwidth comes down to cross the singlet ground state. Therefore, the H_{c1} goes down and the H_{c2} goes up with increasing pressure, indicating the increases in the single-ion anisotropy and the bandwidth of the energy dispersion

curve, the former of which is consistent with the pressure dependence of the single-ion anisotropy as mentioned above.

The transition fields H_{m1} and H_{m2} decrease almost linearly with increasing pressure as shown in Fig. 3. These extremely low transition fields could not be explained by the level crossing of $J = 1$ and $J = 2$ states in a conventional energy scheme of Fe^{2+} ion in Fig. 1(b), and its origin is unknown yet. The pressure dependence of the metamagnetic transition field may be explained by the pressure effect on the single-ion anisotropy D and the single-ion anisotropy D_e in the excited $J = 2$ quintet states as shown in Fig. 1(b). These D and D_e values may change largely by δ/λ where δ is the parameter that describes the splitting of the orbital triplet state $d\epsilon$ by the tetragonal crystalline field as discussed for $FeCl_2 \cdot 2H_2O$ in Fig. 2 of Ref. [3]. To clarify this interpretation, the pressure dependence of the energy difference between $J_1^z = \pm 1$ in $J = 1$ state and $J_2^z = \pm 2$ in $J = 2$ needs to be investigated by INS and ESR measurements.

4 Summary

We performed magnetization measurements of $CsFeCl_3$ at 1.4 K in pulsed high magnetic fields under high pressures. The pressure dependence of H_{c1} is explained fairly by the formula Eq. 2 using the pressure-dependent values of the exchange constants and single-ion anisotropy constant obtained from the INS experiment under pressures. The metamagnetic transition field around 33 T decreases linearly with pressure, which may be explained by the change in the single-ion anisotropy D in the $J = 1$ state and the single-ion anisotropy D_e in the excited $J = 2$ state under pressure.

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