Magnetic structure of pressure-induced ordered state in $CsFeCl_3$

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$CsFeCl_3$ is an easy-plane type antiferromagnet having an integer spin. The crystal structure is a hexagonal with the space group $P6_3/mmc$. At ambient pressure and at zero magnetic field, a singlet ground sate is realized because of the strong easy-plane type single-ion anisotropy [1]. Recent magnetic susceptibility study demonstrated that applying pressure induces a magnetic long-range order [2]. Thus, $CsFeCl_3$ is an interesting compound exhibiting the pressure-induced quantum phase transition. In order to investigate the pressure-induced magnetic long-range order, we performed a single crystal neutron diffraction under pressures at ZEBRA diffractometer in Paul Scherrer Institut (PSI).

A single crystal sample was grown by the vertical Bridgman method. The single crystal was aligned so that the crystallographic *ab* plane was in the horizontal plane. We measured $(h\ k\ 0)$ and $(h\ k\ \pm 1)$ planes. The crystal was installed into the Hard Al-alloy piston cylinder pressure cell in PSI, and the cell was set in a ^{4}He cryostat. We used the tilting mode of 1D detector. Ge(311) monochromator was chosen to obtain the neutrons with the wavelength of 1.178 Å. A collimator of 80′ was installed between the sample and detector.

The pressures were estimated by measurement of the lattice constant of NaCl crystals mounted under the sample. We used the equations of state $V(P, T)$ for NaCl from Ref. [3]. The absolute accuracy of the pressure measurement was estimated to be ± 1 GPa. The obtained lattice constants of NaCl and calibrated pressures are shown in Table 1.

We observe additional peaks at $(h, k, 0) \pm (1/3, 1/3, 0)$ by applying pressure $P = 0.9$ GPa. Figure 1 shows a temperature evolution of the intensity at $(1/3, 1/3, 0)$ and at $P = 1.8$ GPa. The intensity of the peak increases with decreasing temperature, meaning that it is a magnetic Bragg peak. From the indices of the magnetic peaks, it is found that a magnetic propagation vector $\boldsymbol{k}_{\text{mag}}$ is $(1/3, 1/3, 0)$. The temperature evolution of the intensity was measured at several pressures. As a result, it is found that the transition temperature T_{N} increases with increasing the pressures as listed in Table 1.

In analysis of the magnetic structure, we use a representation analysis. The space group $P6_3/mmc$ and the magnetic propagation vector $\boldsymbol{k}_{\text{mag}} = (1/3, 1/3, 0)$ lead to four irreducible representations (IRs) $\Gamma_3 + \Gamma_4 + \Gamma_5 + \Gamma_6$. The basis vectors for Γ_3 and Γ_4 provide an easy-axis type magnetic structure along the *c* axis, whereas those for Γ_5 and Γ_6 provide a coplanar 120° structure in the *ab* plane. As the result of the Rietveld refinement, we find that the magnetic structure for Γ_3 or Γ_6 is the answer. Since $CsFeCl_3$ has the strong easy-plane anisotropy, it is concluded that the structure for Γ_6 is the answer. Thus, the determined magnetic structure exhibits the 120° structure in the *ab* plane.

In summary, the single crystal neutron diffraction under pressure provided the evidence of the pressure-induced magnetic long-range order. Analyzing the magnetic reflections, the magnetic structure is the 120° structure with the propagation vector $\boldsymbol{k}_{\text{mag}} = (1/3, 1/3, 0)$. In addition, the temperature evolutions of the intensities at several pressures shows the pressure dependence of the transition temperature.

References

[1] H. Yoshizawa *et al.*, J. Phys. Soc. Jpn. **49**, 144 (1980).
[2] N. Kurita and H. Tanaka, Phys. Rev. B **94**, 104409 (2016).
[3] J. M. Brown, J. Appl. Phys. **86**, 5801 (1999).

Table 1. Experimentally obtained lattice constant of NaCl and applied pressures at room temperature. Calibrated pressure referred by the table in Ref. [3].

NaCl lattice constant (Å)	Applied P (GPa)	Calibrated P (GPa)	T_N (K)
5.6948 (300 K)	0	-	-
5.5853 (1.6 K)	1.4	0.9	2.8
5.5588 (1.6 K)	1.6	1.3	3.8
5.5491 (1.6 K)	1.8	1.5	4.5
5.5351 (1.6 K)	2.2	1.8	5.5

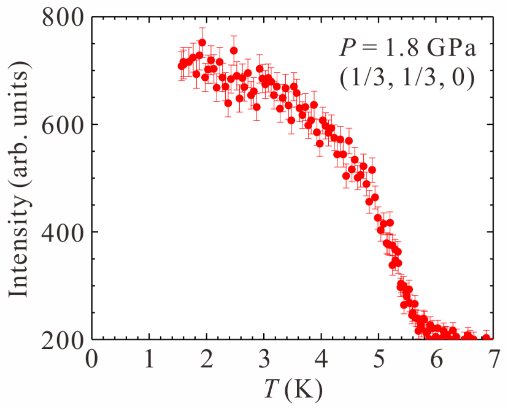

Fig. 1. A temperature evolution of the intensity at $(1/3, 1/3, 0)$ under $P = 1.8$ GPa.