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Multi-step Magnetic Transition in Non-centrosymmetric Compound CeCoGe$_3$

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Abstract. Single crystal neutron diffraction experiments on non-centrosymmetric pressure-induced superconductor CeCoGe$_3$ were carried out in order to reveal a magnetic structure in the ground state. This compound shows multi-step magnetic transitions at $T_{N1} = 20$ K, $T_{N2} = 11.5$ K and $T_{N3} = 7.5$ K. We have observed the appearance of the magnetic reflection at (1 0 1/2) together with the weaker one at (1 0 1/4) and its higher-order harmonics at 2.9 K. These results indicate that the ground state magnetic structure consists of two components with dominant $q_1 = (0 0 1/2)$ and $q_2 = (0 0 3/4)$. No magnetic peak was observed along (0 0 l), indicating that the directions of corresponding antiferromagnetic moments are parallel to the $c$-axis. Consequently, CeCoGe$_3$ has at least two Ce sites in the ground state with respect to the magnitude of the magnetic moment.

1. Introduction

A discovery of the heavy fermion superconductivity in non-centrosymmetric compound CePt$_3$Si opens a new field in condensed matter physics since exotic superconducting properties, such as mixed pairing symmetry, are expected under the lack of inversion symmetry[1, 2]. Intensive research on this subject leads to the finding of a new family CeTX$_3$ where $T$ = transition metal, $X$ = Si, Ge. CeTX$_3$ crystallizes in the tetragonal BaNiSn$_3$-type structure with the space group I4mm. At ambient pressure, CeRhSi$_3$ and CeIrSi$_3$ undergo antiferromagnetic transition at 1.6 K and 5.0 K, respectively, in which the magnetic easy axis is in the basal $c$-plane. Heavy-fermion superconductivity was realized by applying pressure in both compounds. CeRhSi$_3$ becomes superconducting in wide pressure range from 1.2 GPa with the maximum $T_{sc}$ of 1.1 K[3]. Pressure-induced superconductivity in CeIrSi$_3$ appears from 1.8 GPa with relatively high maximum $T_{sc}$ of 1.6 K[4]. The extremely high upper critical field of superconducting phase $H_{c2}$ for $H || c$ and the strong anisotropy were obtained for both compounds.[5–7] This may be relevant to the lack of the inversion symmetry in their crystal structures.

Recently, pressure-induced superconductivity was also found for another member CeCoGe$_3$ with $T_{sc} = 0.7$ K at 5.5 GPa [8]. The recent work on single crystal has revealed that CeCoGe$_3$ exhibits multi-step magnetic transitions at $T_{N1} = 20$ K, $T_{N2} = 11.5$ K and $T_{N3} = 7.5$ K at ambient
pressure\cite{9,10}, whereas earlier study on polycrystalline sample has reported only two transitions at 21 and 18 K\cite{11}. CeCoGe$_3$ has a strong anisotropy; the magnetic easy axis is the tetragonal $c$-axis, which is in contrast to those in CeRhSi$_3$ and CeIrSi$_3$. The superconductivity emerges in the proximity to the critical pressure for the antiferromagnetic ordered state. Therefore, it is of interest to clarify the magnetic interaction in CeCoGe$_3$.

The previous powder neutron diffraction experiments on CeCoGe$_3$ has reported the appearance of the 1 0 1/2 reflection below 20 K\cite{12}. Since the observed peak was only one, the magnetic structure could not be determined. The aim of the present work is to clarify the magnetic structure at the ground state by means of the single crystal neutron diffraction.

2. Experiments
A single crystalline sample of CeCoGe$_3$ was grown by the flux method. Details of sample preparation have been published in Ref.\cite{9}. Neutron diffraction experiments were carried out on the two-axis diffractometer MUSASI and thermal triple-axis spectrometer TAS-2, both installed at the guide hall of the research reactor JRR-3 of the Japan Atomic Energy Agency (JAEA), Tokai, Japan. Neutrons with a wavelength of 2.44 Å were provided by the vertically bent PG(0 0 2) monochromator in MUSASI. In TAS-2, the PG monochromator and analyzer with vertical focusing were used and the spectrometer was fixed to be a elastic condition in a triple-axis mode with $\lambda=2.359$ Å. The collimations of $g$-80'-80'-open for MUSASI and TAS-2, respectively, were employed. The higher order contamination was removed by the PG filter on the incident beam in both instruments. The single crystal was mounted with ($h$ 0 $l$) to be a horizontal scattering plane. The sample was cooled down to 2.8 K by using the standard closed-cycle refrigerator.

3. Result and discussion
Figure 1(a) shows the line scan profile along the (1 0 $l$) in the reciprocal space for CeCoGe$_3$ taken at 2.9 K. A clear superlattice peak was observed at (101/2), which is consistent with the previous work\cite{12}. In addition the additional weak superlattice peak at (101/4) were also found in the present study, indicated by the arrow. Besides, the 101/4 reflection accompanies the higher order harmonics of 103/4 as shown by the dotted arrow. We have observed superlattice peaks at equivalent positions described with the propagation vectors $q_1=(001/2)$ and $q_2=(003/4)$. Figure 1(b) and (c) shows the temperature variation of peak profile at (101/2) and (101/4), respectively, measured at 2.9 K (ground state) and 25.0 K (paramagnetic state). Both peaks disappear with increasing temperature above $T_N$. Based on these facts, these peaks could have a magnetic origin. The inset of Fig. 1(a) shows (0 0 $l$) scan profiles at respective positions to $q_1$ and $q_2$ measured at 2.9 K. Since no trace of magnetic peak was found in the (00$l$) line, the directions of the magnetic moment corresponding to both $q_1$ and $q_2$ are parallel to the tetragonal $c$-axis. This direction of the magnetic moment is consistent with an anisotropy in the magnetization measurement\cite{9}.

In case that some independent magnetic propagation vectors exist at the same temperature, there are several possible models to reproduce the observed results including a volume fraction for respective $q$. Hereafter, we assume a homogeneous magnetic state for CeCoGe$_3$ and try to figure out a possible magnetic structure for the ground state. Since the intensity of (101/2) reflection is 7 times stronger than that of (101/4), the $q_1$ component should predominate the ground state of CeCoGe$_3$. The simple model for the $q_1$ sublattice magnetic structure could be represented by the up-up-down-down sequence along the $c$-direction, as shown in Fig. 2(a). In this model, all Ce ions in the sublattice have the same magnitude of magnetic moment pointing along the $c$-axis. In the following, we try to reproduce the observed magnetic intensity based on this sublattice model. The magnetic scattering intensity for unpolarized neutron is represented
Figure 1. (a) Line scan profile along the (1 0 l) direction of CeCoGe$_3$ measured at 2.9 K. The arrow and dotted arrow indicate the position of (1 0 1/4) and (1 0 3/4) where the weak magnetic peak was observed. The inset shows the (000 l) scan at the same temperature. The $\theta$-2$\theta$ scan profile at (b) (1 0 1/2) and (c) (1 0 1/4) taken at 2.9 K(○, ground state) and 25.0 K(●, paramagnetic state).

as follows:

$$I_{\text{mag}}(Q) \propto |F_{\text{mag}}(Q)|^2 \mu_1^2 f^2(Q)(\sin \alpha)^2 L(\theta)$$  \hspace{1cm} (1)

where $F_{\text{mag}}$ is the magnetic structure factor, $f(Q)$ is the magnetic form factor, $\alpha$ is the angle between the ordered magnetic moment and the scattering vector $Q$, and $L(\theta)$ is the Lorentz factor. The observed magnetic intensity of magnetic reflections with $q_1$ in the ground state is well reproduced as shown in Fig. 2(b). The magnitude of the sublattice magnetic moment $\mu_1$ is deduced to be 0.5(1) $\mu_B$, which agrees with the value expected from the previous work[9].

Although the main component can be described with $q_1$, the ground state of CeCoGe$_3$ incorporates a weak $q_2=(0 0 3/4)$ component in its magnetic structure as well. The existence of the higher-order harmonics for $q_2$ may indicate an anti-phase domain structure; one of the possible configurations is $\uparrow \downarrow \downarrow \uparrow \uparrow \uparrow$ along the $c$-axis. Since the direction of the magnetic moments for both $q_1$ and $q_2$ is parallel to the $c$-axis, the sum of the two components causes the slight oscillation in the size of magnetic moment centered at 0.5 $\mu_B$, in other words, Ce has at least two sites with respect to the magnitude of magnetic moment. It is difficult to find a unique solution for the magnetic structure only from the present neutron diffraction data. Further insights especially form microscopic measurements, like NMR and $\mu$SR are useful to determine the possible ground state magnetic structure, which could also help to discriminate the existence of volume fraction.

In contrast to the simple commensurate antiferromagnetic structure of CePt$_3$Si with $q=(0 0 1/2)$, magnetic structures of CeTX$_3$ family are found to be complicated. The isostructural compound CeRhSi$_3$ is reported to have a magnetic propagation vector $q=\delta 0 1/2$ where $\delta = \pm 0.215$[13]. A longitudinal spin-density wave structure is suggested from the intensity analysis, in which the magnetic moment lies within the $c$-plane.[13] Namely, the size of the magnetic moment is not unique in CeRhSi$_3$ as well. Besides, the magnetic coupling along the $c$-axis is the same as the dominant component $q_1$ in CeCoGe$_3$ whereas the magnetic easy axis is different between these compounds. These characteristic features in the magnetic structure might be common in CeTX$_3$. In order to get further understanding on the magnetic property and its relevance to the non-centrosymmetric structure, detailed temperature dependence in CeCoGe$_3$ as well as the extension to CeIrSi$_3$ are now in progress.
Figure 2. (a) Possible magnetic structure for $q_1$ sublattice in the ground state of CeCoGe$_3$. (b) Calculated and observed magnetic reflection intensity for $q_1=(0 0 1/2)$ with assuming the structure shown in the left panel. The sublattice magnetic moment of Ce is deduced to be $\mu_1=0.5(1)\mu_B$.

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