Unique Magnetic Phases in an Antiferromagnet CeCoGe₃

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We report on the magnetic properties of CeCoGe₃ by measuring the electrical resistivity, magnetic susceptibility, magnetization and specific heat, together with the magnetic properties of a reference compound LaCoGe₃. The measurements were performed on a single crystal grown from a high-temperature solution using a third element, bismuth as flux. From the magnetic susceptibility and magnetization measurements, CeCoGe₃ is an antiferromagnet with a Néel temperature \( T_N = 21 \) K and \( T_N = 8 \) K. The magnetization for \( H \parallel [001] \) indicated a metamagnetic transition with three steps. We constructed a magnetic phase diagram from the magnetization measurements, which is very complex and is roughly similar to the previous result performed on a polycrystalline grain-aligned sample.

KEYWORDS: CeCoGe₃, LaCoGe₃, antiferromagnetism, metamagnetism, magnetization, magnetic resistivity, specific heat.
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1. Introduction

Ternary rare earth germanides crystallize in a variety of crystal structures and have been studied extensively.¹-⁴) Recently we studied the electrical resistivity of an antiferromagnet CeNiGe₃,⁵) which crystallizes in the orthorhombic SmNiGe₃-type structure with the space group \( \text{Cnmm} \). The electrical resistivity under pressure \( P \) revealed that the electronic states of CeNiGe₃ can be tuned by pressure from the Kondo state with antiferromagnetic ordering to the non-magnetic state, crossing the heavy fermion state at a critical pressure \( P_c \approx 5.5 \) GPa. Superconductivity was discovered around the critical pressure.

In continuation to our studies on CeNiGe₃, we investigated the anisotropic physical properties of CeCoGe₃ which has one valence electron less than CeNiGe₃. Unlike CeNiGe₃, CeCoGe₃ crystallizes in the tetragonal BaNiSn₁₂-type crystal structure with lattice parameters \( a = 4.319 \) Å and \( c = 9.829 \) Å and space group \( \text{I4mm} \). The crystal structure of CeCoGe₃ is shown in Fig. 1. The Ce atoms are occupied on the four corners and in the body center of the tetragonal crystal structure, similar to the well-known ThCr₂Si₂-type structure. The nearest neighboring Ce–Ce interatomic distance is \( 4.319 \) Å and the next nearest neighboring Ce atom is \( 5.787 \) Å apart. The present tetragonal crystal structure indicates that the anisotropy may play a crucial role in understanding the magnetic properties. The previous studies on CeCoGe₃ were performed on polycrystalline⁶,⁷) and grain-aligned polycrystalline⁸) samples. The studies on the polycrystalline samples indicated that CeCoGe₃ orders magnetically at temperatures less than 20 K. On the other hand, the reports on the grain-aligned polycrystalline sample indicated two magnetic transitions, one at 21 K and the other at 18 K, corresponding to ferrimagnetic and antiferromagnetic transitions, respectively, and the magnetic isotherms below 20 K exhibited metamagnetic transitions for the grain-aligned direction, parallel to the [001] direction.

In order to investigate the magnetic properties more precisely, we succeeded in growing a single crystal of CeCoGe₃ and studied the anisotropic magnetic properties by means of the electrical resistivity, magnetic susceptibility, magnetization and specific heat measurements. The magnetization measurements were performed precisely in the temperature range from 2 to 24 K, especially to focus a metamagnetic transition with three steps along the [001] direction. An interesting magnetic phase diagram was constructed for CeCoGe₃.

2. Experimental

Single crystals of CeCoGe₃ and LaCoGe₃ were grown by the flux method. Our initial attempt to grow the single crystal of CeCoGe₃ from a stoichiometric melt by the Czochralski pulling method did not yield any single crystals,
indicating the incongruent melting nature of CeCoGe₃. With our earlier success in the growth of single crystals of CePtGe₃ from bismuth as flux,⁹ we used bismuth as flux for the growth of CeCoGe₃. At first, polycrystalline ingots of CeCoGe₃ and LaCoGe₃ were prepared by arc-melting stoichiometric quantities of high-pure metals of Ce and La (99.9%), Co (99.99%) and Ge (99.999%). The polycrystalline samples of CeCoGe₃ and LaCoGe₃, together with appropriate amount of Bi–metal, were placed in alumina crucibles. The crucible was then sealed in a quartz ampoule with a partial pressure of argon gas and placed inside an electric furnace. The temperature of the furnace was raised to 1050°C. After homogenization for about 24 h, the furnace was cooled down to 650°C over a period of two weeks and then finally cooled down to room temperature at a faster rate. The excess bismuth flux was removed by means of centrifuging. The (001) plane-faceted single crystals were then finally cooled down to room temperature at a faster rate.

The crystals were oriented along the principal directions, namely [100] and [001] directions, by means of the Laue back reflection method. Well defined Laue diffraction spots, together with the tetragonal symmetry pattern, indicated the good quality of the single crystals. The crystals were cut along the principal directions using a spark erosion cutting machine for the anisotropic physical property measurements. The magnetization measurements were performed in a commercial SQUID magnetometer, together with a pulsed-field magnetization up to 40 T only for \( H \parallel [001] \). The electrical resistivity was measured using the standard DC four-probe method. The specific heat measurements down to 2 K were performed in a commercial physical property measurement system (PPMS) with the adiabatic heat pulse method.

3. Experimental Results

3.1 Electrical resistivity

The temperature dependence of the electrical resistivity of CeCoGe₃ in the temperature range from 1.3 to 300 K is shown in Fig. 2(a). It can be seen from Fig. 2(a) that the electrical resistivity is highly anisotropic for the current direction \( \mathbf{J} \parallel [100] \) and [001] directions, most likely reflecting the quasi-two dimensional electronic state. The low residual resistivity of \( \rho_0 \) = 0.97 \( \mu \Omega \cdot \text{cm} \) and 2.90 \( \mu \Omega \cdot \text{cm} \) for the current along [100] and [001], respectively, indicates a high-quality sample with the residual resistivity ratio RRR = 124 and 94, respectively. From room temperature to about 150 K, the resistivity remains almost constant and then decreases rapidly. The magnetic ordering is clearly marked by a change of slope in the resistivity at \( T_{N1} \approx 21 \) K for the current along both the directions. This magnetic ordering temperature corresponds well to the previous reports on the polycrystalline samples.⁵⁻⁸ However, for the current along [001], the low-temperature resistivity revealed two more magnetic transitions at \( T_{N2} = 12 \) K and \( T_{N3} = 8 \) K, as shown in Fig. 2(b). These two magnetic transitions are also seen in the magnetic susceptibility measurement, which will be described later.

The temperature dependence of the electrical resistivity of a LaCoGe₃ single crystal for \( \mathbf{J} \parallel [100] \) is shown in Fig. 3. The residual resistivity and residual resistivity ratio are \( \rho_0 = 0.27 \mu \Omega \cdot \text{cm} \) and \( \text{RRR} = 150 \), indicating a high-quality sample. The resistivity of LaCoGe₃ shows a usual metallic temperature dependence. The resistivity data are thus fitted to the Bloch–Grüneisen formula:

\[
\rho = \rho_0 + 4RT \left( \frac{T}{\Theta_D} \right)^4 \int_0^{\Theta_D/T} \frac{x^5 \, dx}{(e^x - 1)(1 - e^x)},
\]

where \( \Theta_D \) is the Debye temperature and \( R \) is a constant and \( \rho_0 \) is the residual resistivity. The fitting parameters are: \( \rho_0 = 0.27 \mu \Omega \cdot \text{cm}, \quad R = 0.14 \mu \Omega \cdot \text{cm/K} \) and \( \Theta_D = 300 \) K. The value of \( \Theta_D \) obtained from the fit is in agreement with \( \Theta_D = 280 \) K obtained from the specific heat data which will be shown later.

The magnetic part of the electrical resistivity \( \rho_{\text{mag}} \) calculated from the difference in the electrical resistivity of CeCoGe₃ and LaCoGe₃ is shown in Fig. 4. With decreasing the temperature, the resistivity increases logarithmically, as expected for a typical Kondo scattering. For temperatures below 100 K, the resistivity falls very rapidly, which may be attributed to the coherence characteristic of a Kondo lattice. The magnetic resistivity \( \rho_{\text{mag}} \) shows a broad
peak at high temperatures due to the interplay between the crystalline electric field (CEF) effect and the Kondo effect, as explained theoretically.\textsuperscript{10,11} The sharp drop in the crystalline electric field (CEF) effect and the Kondo effect, peak at high temperatures due to the interplay between the temperature range. The measurements were carried out with a magnetic field of 2 T and 0.1 T for $H \parallel [100]$ and $H \parallel [001]$, respectively, because the magnetization for $H \parallel [001]$ at 2 K shows metamagnetic transitions at $H = 0.19$ and 0.84 T, as shown by open squares in Fig. 6. At 24 K, the magnetization increases linearly as a function of magnetic field. The magnetization at 18 K, however, indicates a so-called ferrimagnetic curve. Below $T_{N1} = 21$ K, the susceptibility increases rapidly with decreasing temperature. An increase of the susceptibility in Fig. 5 corresponds to the development of a spontaneous magnetic moment, as shown by the magnetization curves at 18, 16 and 14 K in Fig. 6. The temperature dependence of susceptibility for $H \parallel [001]$ clearly shows an inflection point at $T_{N1} = 21$ K, indicating the magnetic ordering.

For further lowering temperature, the susceptibility in Fig. 5 shows two magnetic transitions at $T_{N2} = 12$ K and $T_{N3} = 8$ K, as also observed in the resistivity measurement. At these transitions, the susceptibility shows a step-like decrease with decreasing temperature, which is due to the appearance of metamagnetic transition in a low-field region, as indicated by the magnetization curves at 2 and 10 K in Fig. 6. Here we note that the previous studies on a grain-aligned polycrystalline sample\textsuperscript{8} indicated only two magnetic transitions at about 18 and 21 K, respectively.

The magnetic susceptibility follows the Curie–Weiss law at temperatures larger than 150 K, as shown in Fig. 7. The effective magnetic moment $\mu_{eff}$ and the paramagnetic Curie temperature $\theta_p$ are estimated to be 2.23 $\mu_B$/Ce and $-71$ K, respectively, for $H \parallel [100]$ and 2.16 $\mu_B$/Ce and $-29$ K, respectively, for $H \parallel [001]$. The effective moments are slightly smaller than the free ion moment of Ce\textsuperscript{3+} (2.54 $\mu_B$/Ce). The solid lines in Fig. 7 are the results of the CEF calculations, which are described later.

The typical isothermal magnetization curves for $H \parallel [100]$ and [001] at 2 K are shown in Fig. 8(a). A three-step metamagnetic transition is observed for $H \parallel [001]$: $H_{c1} = 0.19$ T, $H_{c2} = 0.84$ T and $H_{c3} = 3.0$ T. In the previous report on the grain-aligned polycrystalline sample, $H_{c1}$ and $H_{c2}$ were not distinguished each other but $H_{c3}$ was determined correctly. At a field of 7 T, the magnetization for $H \parallel [001]$ amounts only to 0.42 $\mu_B$/Ce, significantly reduced, because the magnetic moment of 1.0 $\mu_B$/Ce is commonly observed in Ce-based ternary intermetallics. We also confirmed from the pulsed-high-field magnetization measurement that there is no metamagnetic transition above $H_{c3} = 3.0$ T, as shown in Fig. 8(b). For $H \parallel [100]$, the magnetization increases linearly up to 7 T and the magnetization at 7 T is found to be 0.15 $\mu_B$/Ce, indicating highly anisotropic magnetizations.

In order to further investigate the metamagnetic transitions, we performed a precise isothermal magnetization
measurement from 2 to 24 K, which is shown in Fig. 9. At 2 K, the metamagnetic transition with three steps is found at $H_{c1} = 0.19$ T, $H_{c2} = 0.84$ T and $H_{c3} = 3.0$ T, as mentioned above. The third metamagnetic transition $H_{c3}$ increases from a field $H_{c3} = 3.0$ T at 2 K to $H_{c3} = 4.1$ T at 16 K and then decreases to lower fields for temperatures above 16 K and finally disappears at 22 K, where the magnetization at higher temperatures increases linearly as a function of magnetic field, indicating the paramagnetic state.

A magnetic phase diagram was thus constructed, as shown in Fig. 10. The characteristic feature of the phase diagram is that the metamagnetic transition with three steps is observed below 8 K and then reduced into two steps above 8 K. The two-step metamagnetic transition is observed in a very narrow temperature range from $T_{N3} = 8$ K to $T_{N2} = 12$ K. The one-step metamagnetic transition is thus observed in the temperature range from $T_{N2} = 12$ K to $T_{N1} = 21$ K, although in this temperature range, CeCoGe$_3$ possesses a ferromagnetic spontaneous magnetic moment at low fields, as shown in Fig. 6.

From these experimental results, it is inferred that CeCoGe$_3$ is an antiferromagnet. The antiferromagnetic easy-axis thus corresponds to the [001] direction (c-axis), while the hard-axis corresponds to the [100] direction (a-axis).

### 3.3 Specific heat

The temperature dependence of the heat capacity $C$ from 2 to 30 K of CeCoGe$_3$ and LaCoGe$_3$ is shown in Fig. 11. The specific heat of LaCoGe$_3$ is small and approximately linear in Fig. 11(b) as expected for a non-magnetic com-

![Fig. 8](image1)

(a) Magnetization at 2 K, measured by the SQUID magnetometer and (b) pulsed-field magnetization at 1.3 K in CeCoGe$_3$. Dashed lines in (b) are the results of CEF calculations.

![Fig. 9](image2)

Fig. 9. Isothermal magnetization in CeCoGe$_3$ for $H \parallel [001]$ at various temperatures.

![Fig. 10](image3)

Fig. 10. Magnetic phase diagram of CeCoGe$_3$ for the field along [001].

![Fig. 11](image4)

(a) Specific heat of CeCoGe$_3$ and LaCoGe$_3$ single crystals. (b) Specific heat in the form of $C/T$ vs $T$. 

![Fig. 12](image5)
From the low-temperature data of LaCoGe$_3$, the $\gamma$ and $\beta$ values in the form of $C = \gamma T + \beta T^3$, which is due to the electronic specific heat $\gamma T$ and the phonon contribution $\beta T^3$, are obtained by the usual method: $\gamma = 4.4$ mJ/(K$^2$.mol) and $\beta = 0.4426$ mJ/(K$^3$.mol). The characteristic Debye temperature $\Theta_D$, calculated from the $\beta$ value, is 280 K which agrees well with the $\Theta_D$ value estimated from the resistivity data as mentioned earlier.

The specific heat data of CeCoGe$_3$ clearly shows a sharp peak at $T_{N1} = 21$ K, indicating the magnetic ordering which was observed in the resistivity and magnetization measurements as mentioned above. However, the specific heat measurement did not show any anomaly at $T_{N2} = 12$ K and $T_{N3} = 8$ K, in contrary to the resistivity and magnetization measurements, although there is found a broad hump around 18 K. The low-temperature specific heat in the form of $C/T$ versus $T^2$ is shown in Fig. 11(b). The $\gamma$ value obtained from the linear extrapolation of the low temperature data is $\gamma = 32$ mJ/(K$^2$.mol). The present $\gamma$ value of CeCoGe$_3$ and that of LaCoGe$_3$ are in good agreement with the previous values.8)

The magnetic contribution to the specific heat, $C_{mag}$ is obtained by subtracting the lattice specific heat of LaCoGe$_3$, $C$(LaCoGe$_3$) from the specific heat of CeCoGe$_3$, $C$(CeCoGe$_3$); $C_{mag} = C$(CeCoGe$_3$) − $C$(LaCoGe$_3$). Figure 12(a) shows the temperature dependence of $C_{mag}$ for temperatures up to 80 K. In addition to a sharp peak corresponding to a magnetic transition, a broad peak centered around 60 K is also observed, which is due to the Schottky anomaly. The magnetic specific heat, $C_{mag}$ consists of $C_{ord}$ and $C_{Sch}$. The former $C_{ord}$ is related to the antiferromagnetic ordering, which is derived from the contribution of the ground state doublet in the CEF scheme, described later. On the other hand, the latter $C_{Sch}$ is derived from the CEF splitting between two excited doublets and the ground state doublet. A solid line in Fig. 12(a) is a calculated Schottky specific heat $C_{Sch}$ based on the CEF scheme, which will be discussed later. The magnetic entropy $S_{mag}$, calculated from the experimental data, is also shown Fig. 12(b). The entropy gain reaches 0.68 ln 2 at $T_{N1}$. The remaining 32% loss of entropy is recovered at about 38 K.

4. Discussion

Here we discuss the crystalline electric field (CEF) state in CeCoGe$_3$. The $J = 5/2$ multiplet of the Ce$^{3+}$ ion splits into following three doublets in the CEF effect with tetragonal symmetry:

$$|\Gamma^{(2)}\rangle = a \left| \pm \frac{5}{2} \right\rangle + b \left| \pm \frac{3}{2} \right\rangle,$$

$$|\Gamma^{(4)}\rangle = a \left| \pm \frac{3}{2} \right\rangle - b \left| \pm \frac{5}{2} \right\rangle,$$

$$|\Gamma_{6}\rangle = \pm \left| \frac{1}{2} \right\rangle.$$

With regard to the CEF scheme in CeCoGe$_3$, here we summarize new findings in the present study as follows. Below $T_{N3} = 8$ K, the magnetization for $H \parallel [100]$ at 2 K increases linearly up to 7 T and a magnetization value at 7 T is about 0.15 $\mu_B$/Ce. On the other hand, the magnetization for $H \parallel [001]$ shows three metamagnetic transitions and nearly saturates around 7 T with a saturation moment of about 0.42 $\mu_B$/Ce. The strong Ising-type anisotropy is characteristic in the ordered state. In addition, the high-field magnetization for $H \parallel [001]$ at 1.3 K revealed that there is no metamagnetic transition above 7 T, and the magnetization is almost constant up to about 40 T, showing a magnetization value of about 0.46 $\mu_B$/Ce. This result indicates that the CEF ground state should be $|\Gamma_6\rangle = \pm \left| \frac{1}{2} \right\rangle$, because this state has $gJ = 0.43$. When $|\Gamma^{(2)}\rangle$ or $|\Gamma^{(4)}\rangle$ becomes the ground state, the magnetization for $H \parallel [001]$ is, simply thinking, expected to increase gradually when the magnitude of magnetization crosses 0.43 $\mu_B$/Ce, and saturates with a magnetization value of 1.29 or 2.14 $\mu_B$/Ce in high magnetic fields.

The second important result is the broad peak observed around 60 K in the temperature dependence of magnetic specific heat. This is due to a Schottky excitation in the CEF scheme and gives useful information about a splitting energy between CEF sublevels. The magnetic entropy reaches about 1.5R ln 2 (= 0.75R ln 4) at 80 K. From these experimental results, it is suggested that the splitting energy between the ground state and the first excited state is larger than 100 K.

In order to analyze these observed experimental data on the basis of a CEF model, we introduce the following Hamiltonian:

$$\mathcal{H} = \mathcal{H}_{CEF} - gJ \mu_B \sum_{\alpha=x,y,z} H_{\alpha} J_{\alpha},$$

$$- \sum_{\alpha=x,y,z} h_{\alpha}^0 J_{\alpha}^0 - \sum_{\alpha=x,y,z} h_{\alpha}^2 J_{\alpha},$$

where $g$ is the Landé $g$-factor (6/7 for Ce$^{3+}$) and $\mu_B$ the Bohr magneton. $\mathcal{H}_{CEF}$ is the CEF Hamiltonian for tetragonal point symmetry and is given as

$$\mathcal{H}_{CEF} = B_{x}^0 O_{x}^0 + B_{y}^0 O_{y}^0 + B_{z}^0 O_{z}^0,$$

where $B_{\alpha}^0$ are the CEF parameters and $O_{\alpha}^0$ the Stevens operators.14,15) The second term in eq. (5) is the Zeeman
The Schottky specific heat, respectively. On the basis of this Hamiltonian, we calculated the temperature dependence of the Schottky specific heat, determined by diagonalizing eq. (5). The magnetic susceptibility was also calculated by

$$\chi^{\mu}_{CEF} = M_\mu / H_\mu$$

with the magnetic field $H = 1$ T. Here we assumed a small temperature-independent susceptibility $\chi_0$ as follows:

$$\chi^{\mu}_B = \chi^{\mu}_{CEF} + \chi^{\mu}_0.$$  

The solid lines in Figs. 7 and 12(a) and dashed lines in Fig. 8(b) are the calculated results by using the CEF parameters listed in Table I. A large molecular field $\lambda^c_1 = 450$ (emu/mol)$^{-1}$ for the $|\pm 1/2\rangle$ ground state is necessary because the ordered magnetic moments of 0.42 $\mu_B$/Ce are directed along the $c$-axis (the [001] direction) and change the magnetic structure in the magnetic field, showing three metamagnetic transitions. On the other hand, a large negative molecular field $\lambda^c_2 = -125$ (emu/mol)$^{-1}$ was introduced to reproduce the small and linear magnetization for $H \parallel [100]$ at 2 K as well as the high-temperature susceptibility data.

Here we note that the Schottky specific heat $C_{Sch}$ in Fig. 12(a) is calculated only on the basis of the CEF scheme, which is not related to the exchange interaction. The Schottky peak around 60 K is well explained by the present CEF scheme in Table I. On the other hand, the Hamiltonian in eq. (5), which includes the CEF scheme and exchange interaction, is applicable to the high temperature magnetic susceptibility and the high-field magnetization. Anisotropy of the magnetic susceptibility and magnetization as well as the value of the magnetization at high fields are also well explained by the present CEF scheme with the exchange interaction. We cannot, however, extend the present model to the antiferromagnetic ordering and the magnetic structure. Here we speculate naively one of the possible magnetic structures in CeCoGe$_3$.

The three-step metamagnetic transition might be simply explained as follows. The ground state at $H = 0$ T is an antiferromagnetic (AF) state with the ordered moment of $M_s = 0.43 \mu_B$/Ce, forming an $\uparrow \downarrow \downarrow$ spin structure. At three transition fields of $H_{c1}$, $H_{c2}$ and $H_{c3}$, the spin structure changes in the sequence of AF $\Rightarrow \uparrow \downarrow \downarrow \uparrow \Rightarrow \uparrow \downarrow \downarrow \Rightarrow F$ (the field-induced ferromagnetic state), showing the magnetization value of 0, $M_s/4$, $M_s/3$ and $M_s$ at each state, as shown in Fig. 13. A solid line is thus a simple magnetization process.
at $T = 0 \text{K}$ and explains the experimental result fairly well. Note that the transition fields in the solid line are taken at the mid-point of the experimental transition fields. In order to theoretically reproduce the magnetization process, a mean field calculation with exchange interactions should be considered. For a more precise analysis on the CEF and the magnetic structure, the neutron scattering experiment is necessary and is left as a future study.

5. Conclusion

High-quality single crystals of CeCoGe$_3$ have been grown by the flux method using bismuth as flux. The resistivity and magnetization measurements indicated a large anisotropy. The magnetization measurement indicated the characteristic metamagnetic transitions and a very unique magnetic phase diagram has been constructed. The large anisotropy in the resistivity and the susceptibility is due to the tetragonal crystal structure of this compound. The antiferromagnetic ordering at $T_{N1} = 21 \text{K}$ is clearly observed in the resistivity, magnetic susceptibility and the specific heat measurements and corroborates well with the previous results$^{6,8}$ on polycrystalline samples. The resistivity measurement for $J \parallel [001]$ indicate two further magnetic transitions $T_{N2}$ and $T_{N3}$ at 12.7 and 8 K respectively which is further substantiated by the low-field susceptibility measurement for $H \parallel [001]$. The magnetization measurement for $H \parallel [001]$ indicate metamagnetic transitions at $H_{c1} = 0.19 \text{T}$, $H_{c2} = 0.84 \text{T}$ and $H_{c3} = 3.0 \text{T}$. The metamagnetic transitions, together with the rapid decrease of the susceptibility, are characteristic of antiferromagnetic behavior in this compound. The magnetization saturates at small fields to a value of 0.42 $\mu_B$/Ce. A simple magnetic structure was proposed to explain the magnetization curve with three metamagnetic transitions, and also the CEF model with the $|\pm1/2|$ ground state was proposed to explain the anisotropy of the magnetization and magnetic susceptibility data.

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