Non-Fermi liquid behavior on heavy-fermion system Ce$_2$Pt$_6$Ga$_{15}$

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Abstract. We have grown single crystalline Ce$_2$Pt$_6$Ga$_{15}$ and investigated the transport and magnetic properties by means of electrical resistivity, magnetic susceptibility, and specific heat measurements. Our results suggest that Ce$_2$Pt$_6$Ga$_{15}$ is characterized as a heavy-fermion system with an electronic ground state of non-Fermi liquid character. The observed logarithmic temperature dependence of $C_4 f / T$ at lower temperature region is a distinctive feature of a non-Fermi liquid state mediated by a two-dimensional antiferromagnetic spin fluctuation. The $C_4 f / T$ reaches as much as about 0.7 J/(mol-Ce)$^{-1}$K$^{-2}$ at 2K.

1. Introduction

In recent years, quantum critical phenomena have been attracting many attentions in the area of the strongly correlated electron systems [1]. In the systems of which electronic ground states are located around the boundary between the magnetic and the nonmagnetic ground states, the temperature dependences of several physical quantities deviate from a Fermi liquid [2,3]. In many cases such phenomena, namely non-Fermi liquid behaviors can be induced by pressure, magnetic field, and changing atomic concentration [4].

Recently, we have succeeded in synthesizing single crystalline Ce$_2$Pt$_6$Ga$_{15}$ through an investigation of the Ga-rich part of the phase diagram of Ce-M-Ga systems by means of the self-flux technique, where M is a transition element. It is reported that Ce$_2$Pt$_6$Ga$_{15}$ crystallizes in the hexagonal Ce$_4$Pt$_{12}$Ga$_{30}$-type structure with the space group $P6_3/mmc$ [5]. This structure has an intrinsic two dimensional character with a large ratio $c/a \sim 4$. The electronic, magnetic, and thermal properties of Ce$_2$Pt$_6$Ga$_{15}$ are nearly unknown.

In this paper, we present the transport, magnetic, and thermal properties of Ce$_2$Pt$_6$Ga$_{15}$ measured in single crystalline samples. We speculate that Ce$_2$Pt$_6$Ga$_{15}$ is characterized as a heavy-fermion system with an electronic ground state of non-Fermi liquid character even at ambient pressure.

2. Experimental

Single crystals of Ce$_2$Pt$_6$Ga$_{15}$ are grown by a so-called self-flux technique. Following the growing procedure, the excess flux of Ga is removed by centrifugation and then washed out in distilled hot water. The resultant crystals are in the form of hexagonal prisms, reflecting their intrinsic crystal structure with typical dimensions of $4 \times 4 \times 10$ mm. The crystallographic characterization is performed by means of the x-ray powder diffraction on crushed pieces of obtained single crystals.
with CuKα radiation at room temperature. All of the obtained diffraction patterns confirmed the hexagonal Ce₄Pt₁₂Ga₃₀-type structure with the space group P6₃/mmc [5]. No traces of impurity phases are detected. The estimated lattice parameters are \( a = 0.4334(2) \) nm and \( c = 1.660(9) \) nm with ratio \( c/a = 3.83 \). The single crystalline nature is confirmed by the back scattering Laue method. The electrical resistivity measurement is performed by the usual four-probe DC method down to 1.5 K. The magnetic susceptibility and magnetization are measured using a commercial SQUID magnetometer (superconducting quantum interference device, Quantum Design) operating in the range of 2-300 K under an applied magnetic field of up to 55 kOe. The measurement of the specific heat is carried out by the relaxation method using the commercial PPMS (physical property measurement system, Quantum Design) operating in the temperature range of 2-300 K.

3. Results and discussion

The temperature dependence of the electrical resistivity \( \rho \) versus temperature \( T \) for Ce₂Pt₆Ga₁₅ measured with current \( j \) along both \( a \)- and \( c \)-directions is shown in Fig. 1. The absolute values of \( \rho \) are quite anisotropic in both crystallographic directions. The \( \rho \) at 300 K for \( c \)-axis (255 \( \mu \Omega \) cm) is larger than that of \( a \)-axis (50 \( \mu \Omega \) cm) by a factor of 5. This is likely because of the quasi-two dimensional character of the structure.

Overall feature of the \( \rho \) is characterized by a typical heavy-fermion system. The \( \rho \) exhibits two broad maximum as often observed in heavy-fermion materials. Negative logarithmic temperature dependence originates from the dominant contribution of the Kondo scattering. A broad high temperature maxima appearing at around \( T_{h\text{max}}^b = 60 \) K is caused by incoherent Kondo scattering under the influence of crystalline electric field (CEF) splitting of the \( 4f \) state of the Ce ion [6]. The lower maximum at around \( T_{l\text{max}}^t = 2.5 \) K may reflect coherent Kondo scattering, which can be taken as a measure of the Kondo temperature \( T_K \) [7]. Below \( T_{l\text{max}}^t \), the \( \rho \) decreases gradually as the temperature decline, but no indications of Fermi liquid character \( \rho \propto T^2 \) are observed down to 1.5 K.

The temperature dependence of the magnetic susceptibility \( \chi \) for Ce₂Pt₆Ga₁₅ measured under an applied magnetic field \( H = 1 \) kOe parallel to both crystallographic axes plotted as a function of logarithmic temperature scale is depicted in Fig. 2. The \( \chi \) reveals the Curie-Weiss behavior for both directions above 100 K. The effective magnetic moment \( \mu_{eff} \) and the paramagnetic

![Figure 1](image1.png)

**Figure 1.** Temperature dependence of the electrical resistivity \( \rho \) of Ce₂Pt₆Ga₁₅ for current \( j \) applied to \( a \)- and \( c \)-directions plotted on a logarithmic temperature scale.

![Figure 2](image2.png)

**Figure 2.** Temperature dependence of the magnetic susceptibility \( \chi \) of Ce₂Pt₆Ga₁₅ measured in a \( H = 1 \) kOe for \( a \)- and \( c \)-axes as a function of \( \log T \).
Curie temperature $\Theta_p$ are estimated to be $2.53 \mu_B$/Ce-ion (2.45 $\mu_B$/Ce-ion) and $-110$ K (36 K), respectively, for $H \parallel a$ (or $H \parallel c$). These estimated values of $\mu_{eff}$ are close to that of free Ce$^{3+}$ ions ($\mu_{eff} = 2.54 \mu_B$/Ce-ion), which indicates that Ce valence in the compound is nearly 3+. The $\Theta_p$ is estimated to be quite negatively large for $H \parallel a$. Such a large negative $\Theta_p$ indicates the presence of the pronounced antiferromagnetic interactions as reported for some Ce-based Kondo lattice systems, e.g., CeRu$_2$Si$_2$ and CeCoIn$_5$ [6,8]. The large deviation from Curie-Weiss law is seen below 50 K, which is likely attributed to the CEF and/or Kondo effects. Below 10 K, the $\chi$ increases monotonically toward lower temperatures showing logarithmic temperature dependence. The observed temperature dependence is not a Fermi liquid like in character, i.e., approximately temperature independent at low temperatures, but a non-Fermi liquid behavior as seen in such as CeCu$_{15.9}$Au$_{0.1}$ and U$_{0.2}$Y$_{0.8}$Pd$_3$ [4,9].

The isothermal magnetization along the $c$-direction increases with increasing in the magnetic field drawing a slight curvature and then reaches 1.2 $\mu_B$/Ce-ion at 55 kOe with saturating behavior. This value is consistent with the expected magnetic moment of the ground state Kramer’s doublet induced by CEF splitting. As for $H \parallel a$, the magnetization increases monotonically and reaches only 0.12 $\mu_B$/Ce-ion at 55 kOe, which value is one order of magnitude smaller than that of the $c$-direction (not shown here). This strong reduction may be caused by the CEF and/or Kondo effects.

The specific heat divided by temperature, $C/T$, versus $T$ is plotted in Fig. 3 for Ce$_2$Pt$_6$Ga$_{15}$ and a nonmagnetic reference material La$_2$Pt$_6$Ga$_{15}$. The $C/T$ of La$_2$Pt$_6$Ga$_{15}$ shows a normal metallic behavior. The low temperature $C/T$ data can be well fitted by the usual expression $C/T = \gamma + \beta T^2$ as shown in the inset of Fig. 3, yielding the Sommerfeld coefficient $\gamma = 9.0$ mJ/(mol-La)$^{-1}$K$^{-2}$ and the phonon contribution $\beta = 1.74$ mJ/(mol-La)$^{-1}$K$^{-4}$ corresponding to the Debye temperature $\Theta_D = 295$ K. As for Ce$_2$Pt$_6$Ga$_{15}$, the $C/T$ shows upturn around 8 K subsequently increases monotonically down to the lowest temperature of 2 K. No signs of long range magnetic order are observed within the measurement range (2-300 K).

We estimate the contribution of 4f electrons to the specific heat $C_{4f}$, by subtracting the specific heat of La$_2$Pt$_6$Ga$_{15}$ with non-f electrons from the measured that of Ce$_2$Pt$_6$Ga$_{15}$. Figure 4 displays temperature dependence of $C_{4f}/T$ plotted against log $T$. A shoulder like peak around
20 K may arise from the splitting of the ground state multiplet of the Ce ions due to the CEF effects. From this temperature, it is suggested that the first excited CEF state is located ∼60 K above the ground state doublet. This result corresponds to the $T^{\text{th}}_{\text{max}}$ appearing in the $T$ dependence of the $\rho$. As the temperature decline, the $C_4f/T$ increases from 15 K and then almost $-\log T$ dependence, non-Fermi liquid behavior is observed. The $C_4f/T$ value reaches as much as about 0.7 J/(mol-Ce)$^{-1}$K$^{-2}$ at 2 K. Such a logarithmic behavior observed in the $C/T$ is a distinctive character of a non-Fermi liquid state mediated by a two dimensional antiferromagnetic spin fluctuation as observed in several Ce-based heavy-fermion compounds close to the magnetic and nonmagnetic ordered ground state [8,9].

The magnetic entropy $S_{4f}$, estimated from the $C_4f$, is also shown by the solid line in Fig. 4. Note that the $S_{4f}$ is calculated by the $C_4f$ data only above 2 K. The $S_{4f}$ releases 9.0 J/(mol-Ce K)$^{-1}$ at 300 K, which is only 60 % of the high temperature limit $R\log6 = 14.9$ J/(mol K)$^{-1}$ expected for a $J = 5/2$ state, where $R$ is a molar gas constant. Therefore, to investigate the ground state properties of Ce$_2$Pt$_6$Ga$_{15}$, more detailed measurements in lower temperature region are required.

4. Summary
We have succeeded in synthesizing single crystals of Ce$_2$Pt$_6$Ga$_{15}$, which crystallizes in the hexagonal system with the space group $P6_3/mmc$. While the $T$ dependence of the $\rho$ is characteristic of typical heavy-fermion systems, both $\chi$ and $C_4f/T$ exhibit $-\log T$ dependence at low temperature region indicative a non-Fermi liquid behavior. The observed logarithmic temperature dependence of $C_4f/T$ is a distinctive feature of a non-Fermi liquid state mediated by a two-dimensional antiferromagnetic spin fluctuation. These results imply the possibility that the electronic ground state of heavy-fermion system Ce$_2$Pt$_6$Ga$_{15}$ is close to the crossover between the nonmagnetic and magnetically ordered ground states even at ambient pressure. To investigate more detailed information about the ground state nature of Ce$_2$Pt$_6$Ga$_{15}$, advanced measurements in the lowest temperature region are currently underway.

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