Specific heat and electrical resistivity at magnetic fields in antiferromagnetic heavy fermion CeAl₂

T. Ebihara, M. Tsuchiya, Y. Saitoh, J. Jatmika, M. Tsujimoto, Y. Shimura, Y. Matsumoto and S. Nakatsuji

1Department of Physics, Shizuoka University, 836 Oya, Suruga Shizuoka Japan
2Institut for Solid State Physics, Kahsiwanoha, Kashiwa Chiba, Japan
E-mail: ebihara.takao@shizuoka.ac.jp

Abstract. We measured temperature dependence of electrical resistivity and the specific heat as a function of magnetic field up to 8 T in CeAl₂. At the metamagnetic transition around 6 T, the coefficient A (ρ ∝ AT²) and Sommerfeld coefficient reduce by 25 %. Such non drastic reduction implies magnetically insensitive phonon could enhance the A and γ in CeAl₂ even in low temperature.

1. Introduction

Cerium or Ytterbium based intermetallic compounds show heavy fermion, unconventional superconductivity, or non-Fermi liquid phenomena at the vicinity of quantum critical point. Such interesting properties emerge at much lower temperature than Debye temperature and are mainly caused by correlation between 4f and conduction electrons. The electronic states of Ce or Yb systems are tunable by alloying, pressure, or magnetic field.

CeAl₂ is a prototype heavy fermion compound ordering antiferromagnetically at 4 K, which crystalizes into the Laves phase MgCu₂-type cubic structure with lattice parameter of 7.70 Å [1]. Below Tₐ at zero magnetic field, the Sommerfeld coefficient (γ) is 135 mJ/K²mol, which is about 40 times larger than γ=3.65 mJ/K²mol of non-f reference material LaAl₂ [2, 3]. CeAl₂ shows metamagnetic transition approximately at 6 T [6, 4, 5]. The Fermi surface is well clarified using de Haas-van Alphen effect above metamagnetic transition at 6 T. In the field induced ferromagnetic state, Fermi surface of CeAl₂ is closely similar to that of LaAl₂. The effective masses (m*’s) of CeAl₂ are in a range from 1.0 to 17 mₑ [3, 7]. Comparing to effective masses of LaAl₂ between 0.2 and 1.7 mₑ, the mass enhancement values range 5 to 10. Although the specific heat of CeAl₂ was measured at magnetic fields up to 5 T, the specific heat measurement is not performed in field induced ferromagnetic state [8]. Therefore, direct comparison between γ and m* is not possible. The electrical resistivity (ρ) shows ρ~ρ₀ + AT² behavior at low temperature in conventional heavy fermion system. The coefficient A is quadratically proportional to the density of state at Fermi energy (D(E_F)) and thus square γ. Systematic measurements of electrical resistivity are not performed yet when crossing metamagnetic transition approximately at 6 T in CeAl₂, although detailed electrical resistivity measurements under high pressure were performed in low temperature [9]. Moreover in low temperature, power law of electrical resistivity deviates from T² as seen in Ref. [10]. CeAl₂ is an exception from conventional heavy fermion.
In this paper, we measured the specific heat and electrical resistivity with crossing the critical field approximately at 6 T for observing specific heat and deviation from standard power laws of electrical resistivity, and for determining the magnetic field dependence of $\gamma$ and $A$ in CeAl$_2$.

2. Experimental Methods

Single crystals of CeAl$_2$ were grown by the Czochralski pulling method. A part of single crystal was milled to fine powder for checking homogeneity, no unfavorable phase and correct composition (i.e. Ce:Al=1:2). Crystallographic directions were determined by X-ray back Laue method and samples were cut by spark cutter. Electrical resistivity was measured by the standard four probe method in the condition of $T>20$ mK and $H<10$ T with dilution refrigerator at Institute for Solid State Physics (ISSP), the University of Tokyo. Specific heat measurement was performed by the relaxation method with dilution refrigerator also at ISSP.

3. Results and Discussion

3.1. Electrical Resistivity

We measured residual resistivity as a function of magnetic field for both $H_\parallel<$100 and $<110$ axes at three temperatures of 100 mK (green), 410 mK (red) and 1000 mK (blue) up to 8 T shown in Figure 1 (a). In Fig. 1 (a), we indicated drops of $\rho_0$ by solid arrows for those axes. Drops for both $H_\parallel<$100 and $<110$ axes are between 5 and 6 T, which reproduces critical magnetic fields determined by magnetization measurements [4, 5, 6]. The samples were polished for enhancing output voltage of measurements. Different amount of defects should be introduced to samples for $H_\parallel<$100 and $<110$ by polishing. Thus, the zero field data are different from each other.

We measured the temperature dependence of electrical resistivity in low temperature as a function of magnetic field. Basically we need to subtract the electrical resistivity of LaAl$_2$ as the phonon part from electrical resistivity of CeAl$_2$. We estimated electrical resistivity of LaAl$_2$ using the Bloch-Grüneisen formula with taking into account the electrical resistivity of LaAl$_2$ seen in Ref. [9]. Electrical resistivity of normal metal is expressed as $\rho=\rho_0+\rho_{ph}(T)$, where $\rho_0$ expresses residual resistivity and $\rho_{ph}(T)$ means scattering by phonon. The $\rho_{ph}$ follows the Bloch-Grüneisen formula in the form of $\rho_{ph}=A'(T/\Theta_D)^5 \int_0^{T/\Theta_D} x^5 \frac{e^x}{(1-e^{-x})(e^x-1)} dx$, where $A'$ is a constant term and $\Theta_D$ means Debye temperature. After we took into account the error bars when reading data from article, we regarded the electrical resistivity of LaAl$_2$ is nearly zero below 20 K. Thus, we show the electrical resistivity in $H<$110 and $H<$100 actually without subtracting electrical resistivity of LaAl$_2$ in temperature below 2 K. In magnetic material, electron-electron ($\rho_{ee}(T)$) and magnetic scattering parts ($\rho_{mag}(T)$) are added to the form to be expressed as $\rho=\rho_0+\rho_{ph}(T)+\rho_{ee}(T)+\rho_{mag}(T)$. The $\rho_{ee}(T)$ is proportional to $AT^2$ in conventional heavy fermion system.

We fitted the electrical resistivity at magnetic fields including zero field from base temperature to 0.4 K by $\rho=\rho_0+A T^2$, where $A$ is the coefficient of quadratic part of electrical resistivity based on the Landau-Fermi-liquid theory. Figure 1 (b) shows electrical resistivity and its fitting curve at low temperature at zero magnetic field. The zero field date are the only exception that we could fit by quadratic function in electrical resistivity of CeAl$_2$. Thus, we used a fitting function as $\rho=\rho_0+A'T^n$ at magnetic fields for both $H_\parallel<$100 and $<110$ axes, where $n$ is $n^{th}$ power and $A'$ is the coefficient of $n^{th}$ power. We summarized $n$ and $A'$ in Fig. 3 (a) and (b).

The values of $n$ are almost constant near $n=3$, which is different from conventional heavy fermion showing $n=2$ as seen in Fig. 3 (a). Such deviation of $n$ from two implies additional factor exists in low temperature electrical resistivity in CeAl$_2$. On the other hand, we could not
Figure 1. (a) Magnetic field dependence of residual resistivity ($\rho_0$) at three temperatures. Open circles for $H \parallel <100>$, closed circles for $H \parallel <110>$, and arrows for indicating critical fields for both axes. (b) Temperature dependence of residual resistivity ($\rho_0$) at zero field. Black open circles show data points and a blue line represents quadratic fitting in a range from base temperature to 0.4 K.

Figure 2. Magnetic field dependence of electrical resistivity when (a) $H \parallel <100>$ and (b) $H \parallel <110>$.

see significant change in $n$-value at critical fields. The $A^*$-value should correspond to $A$-value, which is proportional to $D(E_F)$. There are shoulders near critical field seen in Fig. 3 (b).

3.2. Specific Heat
We performed specific heat measurements in $H \parallel <110>$ and $<111>$ in CeAl$_2$ shown in Fig. 4 (a) and (b). Below 0.2 K at magnetic fields, specific heat rapidly increases in magnetic field. The nuclear Schotkey anomaly causes this increase of specific heat. The specific heat is expressed as a function of $C(T) = C_{el} + C_{ph} + C_{nuc}$: $C_{el} = \gamma T$, $C_{ph} = \beta T^3$ and $C_{nuc} = \alpha T^{-2}$. At zero field, $C(T) = C_{el} + C_{ph}$ is used for fitting function because nuclear part does not contribute to the
Figure 3. Magnetic field dependence of (a) $n^{\text{th}}$ power and (b) coefficient $A^*$ for $H \parallel <100>$ and $<110>$ in fitting function; $\rho = \rho_0 + A^* T^n$.

Figure 4. Magnetic field dependence of specific heat when (a) $H \parallel <110>$ and (b) $H \parallel <111>$. 

total specific heat $C(T)$. On the other hand, we use $C(T) = C_{\text{el}} + C_{\text{ph}} + C_{\text{nuc}}$ for fitting data at magnetic fields with three parts. Both results at zero field and at magnetic fields are well fitted by the functions. Examples of fitting at 0 and 3 T are shown in Fig. 5. We determined $\gamma$ and $\beta$ as a function of magnetic field and depicted in Fig.6 (a) and (b). The $\gamma$ and $\beta$ reduce their values about 25 % and 70 % when crossing critical field about 6 T. This result infers there remains magnetically less sensitive enhancement factor in $\gamma$.

If we assume ideal condition such as spherical Fermi surface and electrons travel on the spherical Fermi surface, we can estimate the $\gamma$ from $m^*$. Based on the free electron model, approximately 100 mJ/K$^2$mol is estimated as the $\gamma$-value from $m^*$ about 16 $m_e$ determined by de Haas-van Alphen above 6 T [3, 7]. Therefore, the less reduction of $\gamma$ and $m^*$ implies that magnetically less sensitive enhancement factor may exist around metamagnetic transition. At much higher fields in pulse magnet, Pricopi et al. performed dHvA experiments to observe light effective mass about 3 $m_e$ at 53 T in CeAl$_2$, which is almost the same as that in LaAl$_2$ [11, 12].
Figure 5. Fitting to the specific heat (a) at zero field and (b) at 3 T.

Figure 6. Magnetic field dependence of (a) electronic specific heat coefficient $\gamma$ and (b) lattice specific heat coefficient $\beta$.

In conclusion, we measured electrical resistivity and specific heat at magnetic fields for observing magnetic field dependence of the coefficients $A$ and $\gamma$. We reproduced $T^3$ dependence in electrical resistivity in low temperature. The $\gamma$-value reduces about 25% when crossing metamagnetic transition, but the value maintain large $\gamma$-value as 100mJ/K$^2$ mol. Such non-drastic reduction implies magnetically insensitive phonon could enhance the $A$ and $\gamma$ in CeAl$_2$ even in low temperature. At much higher magnetic fields, we may observe an anomaly in specific heat, magnetostriction or magnetization measurements.

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