Crystal Field Effect on Specific Heat and Magnetic Susceptibility of Single Crystals of Ce\textsubscript{x}La\textsubscript{1-x}Pt\textsubscript{3}Si

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Abstract. Measurements of the temperature dependence of magnetic susceptibility and specific heat down to $\sim 1.5$ K were carried out to investigate the crystalline electric field (CEF) state of CePt\textsubscript{3}Si. In the Ce-dilute region ($x = 0.05$), a single crystal could be prepared successfully by the Czochralski pulling method. The results in dilute alloys indicate that the first excited CEF level is well separated from the ground state doublet; the CEF level schemes with doublets lie at 0, 139, and 176 K.

1. Introduction

CePt\textsubscript{3}Si exhibits antiferromagnetic order at $T_N = 2.2$ K and superconducting transition at $T_c = 0.75$ K[1]. The electronic specific heat coefficient $\gamma$ is large, approx. 0.39 J/mol K\textsuperscript{2}. This compound has a characteristic crystal structure, that is, the lack of inversion symmetry (space group $P4mm$). This characteristic is unique to heavy fermion superconductors.

Metoki et al. performed inelastic neutron scattering experiments on a polycrystalline sample of CePt\textsubscript{3}Si[2]. They observed two peaks at $\Delta E \approx 1$ and 24 meV in the low-$Q$ data of the inelastic neutron scattering spectra and explored the consequences of attributing them to CEF excitations. Takeuchi et al. measured temperature dependences of magnetic susceptibility, specific heat, thermal expansion, and magnetization using single crystal samples, and explained their results on the basis of Metoki’s CEF model[3]. On the other hand, Bauer et al. performed inelastic neutron scattering experiments on a polycrystalline sample and suggested a model different from Metoki’s CEF model, namely, CEF level schemes with doublets lie at 0, 152, and 232 K[4]. Laumann et al. measured the specific heat of CePt\textsubscript{3}Si and Ce\subscript{0.2}La\subscript{0.8}Pt\textsubscript{3}Si and compared experimental results with calculated data[5]. By their estimation of the magnetic entropy, Bauer’s CEF model is good accordance than Metoki’s CEF model.

2. Experimental methods

A series of polycrystalline Ce\textsubscript{x}La\textsubscript{1-x}Pt\textsubscript{3}Si samples were synthesized by arc-melting, the details of which are described in refs. 6 and 7. Single crystals were grown by the Czochralski pulling method using a laboratory-made tri-arc furnace. Chemical composition $x$ of Ce\textsubscript{x}La\textsubscript{1-x}Pt\textsubscript{3}Si is the nominal composition determined from the starting material. X-ray powder diffractions of Ce\textsubscript{x}La\textsubscript{1-x}Pt\textsubscript{3}Si samples were measured to characterize these samples. Although no X-ray powder diffraction patterns are shown here, no extra phases were observed and all of full widths at half maximum of Ce\textsubscript{x}La\textsubscript{1-x}Pt\textsubscript{3}Si peaks were almost unchanged regardless of $x$. Moreover,
lattice constants $a$ and $c$ changed linearly with $x$. Therefore, those of quality is equality at all regions of $x$. The direction of the single crystal was determined from the X-ray Laue pattern.

The temperature dependence of magnetic susceptibility was obtained from dc magnetization by dividing by the external magnetic field $H = 5000$ Oe. The dc magnetization was measured with a SQUID magnetometer in the temperature range of $\sim 1.7$ K to 300 K. Specific heat measurements were conducted by the standard adiabatic heat-pulse method in the temperature range of $\sim 1.5$ K to 20 K.

Figure 1. Temperature dependence of magnetic susceptibility of Ce$_{0.05}$La$_{0.95}$Pt$_3$Si. Full circles (•) indicate magnetic susceptibility for $H \parallel a$ and open ones (○) for $H \parallel c$. Full (——) and broken (– – –) lines indicate calculated results of magnetic susceptibility for $a$- and $c$-axes using the single ion CEF model, respectively. Inset shows the temperature dependence of the inverse of magnetic susceptibility.

3. Results

Figure 1 shows the temperature dependence of magnetic susceptibility $\chi(T)$ with 5000 Oe magnetic fields applied parallel to the $a$- and $c$-axes of a Ce$_{0.05}$La$_{0.95}$Pt$_3$Si single crystal as-grown sample. The inset shows the temperature dependence of the inverse of magnetic susceptibility. An anisotropic behavior is clearly observed below 30 K, and $\chi_a$ at the lowest temperature is two times larger than $\chi_c$ at the same temperature. $\chi(T)$ of the $a$- and $c$-axes above 100 K follow the Curie-Weiss law, but the estimated effective moments are somewhat different from the theoretical value of $2.54 \ \mu_B/\text{Ce}$ for Ce$^{3+}$; $\mu_{\text{eff}}$ for $a$-axis is $2.33 \ \mu_B/\text{Ce}$ and $\mu_{\text{eff}}$ for $c$-axis is $3.83 \ \mu_B/\text{Ce}$. This difference may be caused by background subtraction and a small change in Ce concentration. The chemical composition of Ce$_{0.05}$La$_{0.95}$Pt$_3$Si is the nominal composition determined from the starting material. The change of $x$ for the single crystal sample may not be negligible which was prepared by the Czochralski pulling method. We did not subtract the magnetic susceptibility of LaPt$_3$Si because we could not prepare a single crystal of LaPt$_3$Si. Full and broken lines indicate calculated results of magnetic susceptibility for the $a$- and $c$-axes.
using the single ion CEF model. We consider that the overall feature is described well by our calculation.

Table 1. CEF parameters, energy level schemes, and corresponding wave functions of CePt$_3$Si.

| CEF parameter | $B_{20}$ (K) | $B_{40}$ (K) | $B_{44}$ (K) | $\lambda$ (mol emu$^{-1}$) |
|---------------|--------------|--------------|--------------|----------------------------|
|               | 0.80         | 0.34         | 2.8          | -26                        |

Energy level (K) and wave function

- 176: $0.87 \ket{\pm 5/2} + 0.49 \ket{\mp 3/2}$
- 139: $\ket{\pm 1/2}$
- 0: $0.49 \ket{\pm 5/2} - 0.87 \ket{\mp 3/2}$

We calculated the magnetic susceptibility using the following CEF Hamiltonian in addition to a conventional Zeeman energy term,

$$H_{CEF} = B_{20}O_2^0 + B_{40}O_4^0 + B_{44}O_4^1,$$

which is appropriate to the local symmetry of 4mm at Ce ion site, where $B_{lm}$ and $O_m^l$ are CEF parameters and Stevens operators, respectively. The CEF parameters, energy level schemes, and wave functions are listed in Table 1.

Figure 2. Temperature dependence of the magnetic part of specific heat $C_m$ of annealed polycrystalline Ce$_x$La$_{1-x}$Pt$_3$Si, $x = 1$, 0.5, and 0.2, and single crystal as-grown sample of Ce$_{0.05}$La$_{0.95}$Pt$_3$Si, plotted as $C_m/T$ on a logarithmic temperature scale. Broken (---) and chain (— • —) lines indicate calculated results with CEF splitting as 0-139-176 K and a Kondo contribution with $T_K = 4$ and 1 K, respectively.
Figure 2 shows the temperature dependence of the magnetic part of the specific heat $C_m$ from $\sim 1.7$ K to 20 K, where $C_m$ was obtained by subtracting the specific heat of LaPt$_3$Si from that of Ce$_x$La$_{1-x}$Pt$_3$Si. In the dilute region ($x = 0.2$ and 0.05), calculations with CEF splitting as 0-139-176 K and a Kondo contribution with $T_K = 4$ and 1 K are indicated, respectively. At high temperature ($\sim 20$ K), a increase of $C_m$ with increasing $T$ was observed in the case of dilute alloys. At low temperature, $C_m(T)$ increased with decreasing $T$. This increase of the dilute $x$ is explained well by the $T$ dependence of $C_m$ of Kondo effect. Although we conducted measurements only down to $\sim 1.5$ K, if $C_m(T)$ was extrapolated to low temperature, the integrated entropy up to 20 K would be nearly $R \ln 2$. Therefore, we attribute the higher anomaly to the Schottky anomaly due to the first exciting doublet of CEF level scheme, and the lower anomaly to the anomaly of the Kondo effect. The increase at low temperature with decreasing $T$ shifts to lower with decreasing Ce concentration.

4. Summary

Finally, the magnetic properties of the dilute alloy of Ce$_x$La$_{1-x}$Pt$_3$Si are explained by the CEF model, and we conclude that CEF energy level schemes with doublets lie at 0, 139, and 176 K. Although our specific heat results for Ce$_x$La$_{1-x}$Pt$_3$Si were almost in accordance with previous results of refs. 5 and 8, however, Bauer et al.’s model could not explain our magnetic susceptibility results. On the other hand, magnetic susceptibility results have shown a marked contrast between the magnetic easy axis of the a-axis of our study and that of previous results in refs. 2 and 3. It is the c-axis for CePt$_3$Si single crystal. At present, we cannot explain the difference in magnetic easy axis.

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