Transverse Magnetoresistance of α -Ce

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We have studied the Fermi surface of α -Ce by measuring the transverse magnetoresistance. A high quality single crystal of α -Ce with the residual resistivity of 0.7 $\mu\Omega$ cm was successfully obtained by using the tetra-arc Czochralski and the solid-state electro-transport methods. The results suggest that α -Ce is a compensated metal with electrons and holes of equal number.

KEYWORDS: α -cerium, $\gamma \rightarrow \alpha$ transition, magnetoresistance, Fermi surface

§1. Introduction

The Ce metal undergoes an isostructural $\gamma \rightarrow \alpha$ (fcc \rightarrow fcc) transition with increasing pressure or decreasing temperature. The volume contracts about 16% with this first order transition.

Though many investigations were performed during a 1970's, few reliable data on the physical decade in properties of each phase exist. It is because the high quality individual phases were hardly obtained. In Fig.1, we show the non-equilibrium phase diagram in a low pressure and low temperature region of Ce.1) The solid and dashed lines represent the phase boundaries in cooling and heating processes, respectively. The y-Ce transforms incompletely to a double hexagonal phase β -Ce upon the cooling below 250K at ambient pressure. This β phase remains at low temperature after $\gamma \rightarrow \alpha$ transition at about 100K. The β -Ce is antiferromagnetic below the Néel temperature of 12.5K.2) The small amount of residual β -Ce masks the intrinsic properties of the α -Ce.

The magnetic susceptibility of γ -Ce obeys the Curie-Weiss law.^{3,4)} Burgardt et al. suggested that the resistivity of γ -Ce exhibits Kondo like behavior.²) Recently, Naka et al. measured the pressure effect of the magnetic susceptibility.4) They estimated the Kondo temperature of γ -Ce to be 25K at ambient pressure, assuming the single-impurity Kondo model. The first reliable susceptibility data of α -Ce was given by Koskimaki and Gschneidner.⁵⁾ They indicated that α -Ce is essentially an enhanced Pauli paramagnet.⁵) The susceptibility is 5.3×10^4 emu/mol around 50K, being in agreement with the data by Naka et al.⁴⁾ They also measured the specific heat and gave the electronic specific heat coefficient to be 12.8 mJ/mol.K². These data by Koskimaki and Gschneidner seem reliable because there are no evidence for an anomalous peak near 13K arising from remaining β -Ce.

All these experimental results seem to indicate that the 4f electron in γ -Ce is localized, and transferred to the itinerant electron with $\gamma \rightarrow \alpha$ transition. The γ -Ce will be categorized into the Kondo regime, and α -Ce, into the valence fluctuation regime. The recent photoemission study also suggests that the 4f states in α like Ce is well

characterized by a band-like description.⁶) It is interesting to know the Fermi surface of α -Ce. For this purpose, we investigated the high field transverse magnetoresistance of α -Ce single crystal.

§2. Sample Preparation and Evaluation

We made a single crystal of Ce metal (α -Ce) by the tetraarc Czochralski method. The single-crystal rod was purified by use of the solid-state electro-transport (SSE) method, holding the rod at a temperature of 700°C using a DC



Fig.1 Non-equilibrium phase diagram of Ce. The solid and dashed lines represent the phase boundaries in cooling and heating processes, respectively.¹) The circles and triangles are the points obtained in the present measurement of the resistivity. The thick line shows the process used in the present work. The specimen of α -Ce used in the measurement of the magnetoresistance were obtained after this process.



Fig.2 (a) is the electrical resistivities at a pressure of 0.5GPa and at ambient pressure. The arrows correspond to the starting points of $\gamma \rightarrow \alpha$ and $\alpha \rightarrow \gamma$ transitions. (b) is the curves at the low temperature region for three different specimens. SSE means that the purified specimens by a solid-state electro-transport method was used.

current flow for 4 days in vacuum of 1×10^{-9} torr. To obtain a pure α -Ce, the specimen (γ -Ce) was compressed to 0.5GPa at room temperature and then slowly cooled down to liq. He temperature, following the thick line in the diagram of Fig.1. The medium, commercially named DAPHNE 7373, was used for hydrostatic pressurizing. The specimen was fitted into a teflon cell with a diameter of 5.5mm and pressurized in a piston-cylinder type clamp made of BeCu.

The quality of the specimen $(\alpha - Ce)$ was evaluated by electrical resistivity measurements. Fig.2 shows the the temperature dependence of the electrical resistivity for the [110] direction at a pressure of 0.5GPa and at ambient pressure, OGPa. We can see the abrupt change of the resistivity caused by $\gamma \rightarrow \alpha$ and $\alpha \rightarrow \gamma$ transitions. The starting points of $\gamma \rightarrow \alpha$ and $\alpha \rightarrow \gamma$ transitions are showed by the arrows in Fig.2(a). These temperature are plotted in Fig.1 by the circles and triangles, respectively. These points are in agreement with the previous data showed by the solid and dashed lines in Fig.1.1) Fig.2 (b) shows the resistivity in the low temperature region of three different specimens, where SSE means that the specimen was purified by the SSE method. As shown in Fig.2 (b), the residual resistivity was 2.9 $\mu\Omega$ cm for the specimen cooled at 0GPa and 0.7 $\mu\Omega$ cm at 0.5GPa. The temperature dependence of the resistivity at 0GPa shows a broad swell at temperatures around 13K. This temperature corresponds to the Néel temperature of β -Ce. Thus the small swell is most likely attributable to the antiferro-magnetic ordering of the residual β -Ce in the specimen. The resistivity at 0.5GPa shows no anomaly, indicating that little β -Ce remains in the specimen. By the SSE purification, the residual resistivity was lowered from 1.2 $\mu\Omega$ cm to 0.7 $\mu\Omega$ cm, the smallest ever reported.⁷⁻¹⁰

§3. Results and Discussions

The magnetoresistance has been measured at 1.5K with applied fields up to 12T. Fig.3 shows the transverse magnetoresistance $\Delta\rho/\rho_0$ as a function of the fields in the fundamental directions in the (110) plane with a current along the [110] direction. In the figure, $\Delta\rho=\rho_{\rm H}-\rho_0$, $\rho_{\rm H}$ is the resistivity in an applied field H, and ρ_0 is the residual resistivity. In Fig.3, it can be found that $\Delta\rho/\rho_0$ increases with increasing field, being proportional to H^{1.8} and not saturated for the all of the three field directions [001], [111] and [110]. No anisotropic behavior also can be seen. The inset of Fig.3 is a log-log plot of $\Delta\rho/\rho_0$ vs. H. The value of $\Delta\rho/\rho_0$ is 2.5 at 12T. This value indicates that the high field condition $\omega_c\tau>>1$ is nearly satisfied in the present measurement, where ω_c is the cyclotron frequency and τ is the collision



Fig.3 The transverse magnetoresistance $\Delta \rho / \rho_0$ of α -Ce as a function of the field. The current is along the [110] direction, the temperature is 1.5K and the pressure is 0.5GPa. The inset is a log-log plot of $\Delta \rho / \rho_0$ vs. H.

time of the conduction electron. The value of $\Delta \rho / \rho_0$ at ambient pressure was only 0.6, which doesn't satisfy the high field condition.

The magnetoresistance in the high field limit exhibits a field dependence being proportional to H² in the following two cases; the numbers of electrons and holes are equal, i.e. compensation; and secondly, the Fermi surface is multi-connected and an open orbit exists nearly in the current direction. In the face-centered-cubic structure of α -Ce, there is no open orbit in the field directions [001] and [111], because of the fourfold and threefold crystal symmetries, respectively. Thus the non-saturated behaviors of $\Delta\rho/\rho_0$ for the field along the [001] and [111] directions are presumably caused by the compensation; the numbers of the electrons and holes are equal.

The Fermi surface of α -Ce was calculated by Higuchi and Hasegawa within the local-density approximation framework for the atomic configuration of 5d²6s²4f⁰.¹¹) Their primary calculation indicates that α -Ce is semimetallic with the low carrier concentration of 0.12 per atom. This theoretical result is consistent with the present result of the magnetoresistance, which indicates that α -Ce is an compensated metal.

The present result indicates that α -Ce possesses the valence electrons of even number, because the fcc structure has one atom in an unit cell. It seems the 4f electron in Ce atom becomes to be itinerant in α -Ce. Further, the effective mass of the conduction electron is slightly enhanced in α -Ce, as the electronic specific heat constant is 12.8 mJ/mol.K^{2.5}) In order to obtain the knowledge of the Fermi surface and the effective mass of the conduction electron in α -Ce, the measurement of the de Haas-van Alphen effect should be carried out.

In conclusion, we have successfully obtained a highquality single crystal of α -Ce at low temperature. Highfield transverse magnetoresistance indicates that α -Ce is a compensated metal, and the f electron in α -Ce becomes to be itinerant.

Acknowledgement

This work was supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Science and Culture.

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