Pressure Effects on Electrical Resistivity of Heavy-Fermion Antiferromagnet Ce$_2$PdGa$_{12}$

S Ohara$^1$, T Yamashita$^1$, T Shiraishi$^1$, K Matsubayashi$^2$ and Y Uwatoko$^2$

$^1$ Department of Engineering Physics, Electronics and Mechanics, Graduate School of Engineering, Nagoya Institute of Technology, Nagoya 466-8555, Japan
$^2$ Institute for Solid State Physics, The University of Tokyo, Kashiwa 277-8581, Japan
E-mail: ohara.shigeo@nitech.ac.jp

Abstract. We have succeeded in growing high purity single crystalline Ce$_2$PdGa$_{12}$ which crystallizes in the tetragonal system (space group P4/nbm). Ce$_2$PdGa$_{12}$ is a typical heavy-fermion antiferromagnet with the magnetic ordering temperature of 11K at ambient pressure. We investigate the pressure effects on the electrical resistivity for Ce$_2$PdGa$_{12}$ up to 8GPa. With increasing pressure, the ordering temperature is dramatically suppressed to 3K at 3GPa. The electronic state of Ce$_2$PdGa$_{12}$ are tuned by pressure from the heavy-fermion antiferromagnetic state to the non-magnetic state through a critical pressure around $P_c \sim 7$GPa.

1. Introduction

Many Ce-based intermetallic compounds have been noticed owing to their attractive electronic properties, such as unconventional heavy-fermion superconductivity and magnetic transition [1]. Pressure can change the magnetically ordered ground states of Ce-based systems to non-magnetic one. Furthermore, in some cases anomalous phenomena such as non-Fermi liquids and heavy-fermion superconductivity appear in the vicinity of a quantum critical point at which the magnetic order is destroyed [2]. Many heavy-fermion superconductor, including pressure-induced one, have been reported for the Ce-based compounds which crystallizes in a tetragonal symmetry, for example, CeCu$_2$Si$_2$ [3] and Ce$_n$MIn$_{3n+2}$ (n=1,2; M=Co, Rh) [4, 5, 6, 7]. Therefore, the interest in Ce-based tetragonal materials has been stimulated.

Ce-based ternary compounds Ce$_2$MGa$_{12}$ (M=Ni, Pd) have been reported as heavy-fermion antiferromagnets with Neel temperature $T_N = 10$K and 11K for $M=$Ni and Pd, respectively [8, 9]. These compounds crystallize in the tetragonal system (space group P4/nbm) [8]. The structure can be viewed as Ce atoms-layer contained in Ga cavities alternating with Ga segments and MGa$_6$ segments along the crystallographic c-axis [8]. The Ce atom is arranged in a two-dimensional square lattice site perpendicular to the c-axis.

Recently we have succeeded in growing the single crystalline Ce$_2$MGa$_{12}$ (M=Ni, Pd, Pt). We have found that Ce$_2$PtGa$_{12}$ is also a heavy-fermion antiferromagnet with the magnetic ordering temperature of 7 K at ambient pressure. For Ce$_2$PdGa$_{12}$, we obtained clean sample with the residual resistivity ratio $\sim 50$. Here, we present the first measurements of the pressure effects on the electrical resistivity of Ce$_2$PdGa$_{12}$. 

Published under licence by IOP Publishing Ltd
2. Experimental

Single crystals of $\text{Ce}_2\text{PdGa}_{12}$ is synthesized in Ga-flux. Ce ingot (3N), Ni ingot (5N) and Ga ingot (5N) are placed into an alumina crucible in a 2:1:40 ratio. The alumina crucible is sealed in a quartz ampoule under high vacuum. The ampoule is heated up to 800$^\circ$C over 6 hours, then held for 5 hours and last, arrowed to cool slowly to 300$^\circ$C at a rate of 5$^\circ$C/h. The excess Ga surrounding the grown crystals is removed using centrifugal force and then wash out in distilled hot water. Crystallographic characterization is performed by means of X-ray powder diffraction with Cu$K_\alpha$ radiation. The X-ray powder diffraction pattern of $\text{Ce}_2\text{PdGa}_{12}$ is in good agreement with that of tetragonal Sm$_2\text{PdGa}_{12}$ type structure. The obtained lattice parameter are $a=0.6111(2)$ nm $c=1.5558(6)$ nm. These values are slightly large compared to those of previous report [8]. The electrical resistivity are measured in the temperature range 2-300 K using the conventional DC four-proved method with the current parallel to $a$-axis. The pressure experiments on the electrical resistivity are done using a cubic-anvil cell up to 8 GPa.

3. Results and Discussion

Figure 1 shows the logarithmic scale of temperature dependence of the electrical resistivity for $\text{Ce}_2\text{PdGa}_{12}$ and a non-magnetic reference material of $\text{La}_2\text{PdGa}_{12}$ at ambient pressure. $\text{La}_2\text{PdGa}_{12}$ shows superconducting transition at 2.7 K. The electrical resistivity of $\text{Ce}_2\text{PdGa}_{12}$ corresponds to the typical heavy-fermion features with a antiferromagnetic ordering temperature of $T_N=11$K. The magnetic contribution to resistivity for $\text{Ce}_2\text{PdGa}_{12}$ is obtained by subtracting the phonon term of the resistivity of $\text{La}_2\text{PdGa}_{12}$. The magnetic contribution of $\rho_m$ is shown by broken line in Fig. 1. $\rho_m$ has a broad maximum around $T_{CEF}=50$K and decreases steeply below $T_N$. The broad maximum is due to a Kondo scattering of the conduction electron at the first excited crystalline electric field (CEF) state. The strong reduction of the magnetic scattering below $T_N$ is attributed to the antiferromagnetic ordering. Below $T_N$, the resistivity is proportional to $T^2$.

Figure 2 shows $\rho_m$ vs. log $T$ plot for several pressures. With increasing pressure, the electrical resistivity of $\text{Ce}_2\text{PdGa}_{12}$ increases in magnitude in the high temperature region. The coefficient of -log$T$ dependence above $T_{CEF}$ is clearly enhanced by pressure. The $T_{CEF}$ decreases up to 6GPa and then increases at 8GPa. To clarify the pressure effects on the characteristic temperature, we show $\rho_m$ at low temperature region in a linear scale in Fig. 3. The antiferromagnetic ordering temperature of $T_N$ dramatically suppressed from 11K at ambient pressure to 3K at 3GPa. From 2 to 6 GPa the resistivity shows a shoulder. The shoulder or maximum which is observed in many heavy-fermion compounds at low temperature scales with the Kondo temperature[10]. From the resistivity data, we could not assign $T_{shoulder}$

![Figure 1. Logarithmic scale of temperature dependence of the electrical resistivity for $\text{Ce}_2\text{PdGa}_{12}$ and $\text{La}_2\text{PdGa}_{12}$ at ambient pressure. The broken line shows the magnetic contribution of $\rho_m$ for the resistivity of $\text{Ce}_2\text{PdGa}_{12}$ (see text). Characteristic temperatures $T_{CEF}$, $T_N$ and $T_c$ are described in the text.](image-url)
above 4GPa to any magnetic order, or to the Kondo coherent temperature. However, at 8GPa the shoulder disappears or merge with the maximum at $T_{CEF}$. The overall feature of resistivity at 8GPa is generally attributed to a crossover from incoherent scattering of electrons at high temperature to the development of Kondo coherent states at low temperature (see Fig. 2). Thus we conclude that the electronic state of Ce$_2$PdGa$_{12}$ are tuned by pressure from the heavy-fermion antiferromagnetic state to the non-magnetic state through a critical pressure around $P_c$~7GPa. The pressure effects on the resistivity of Ce$_2$PdGa$_{12}$ are quite similar to those observed in a

Figure 2. Logarithmic scale of temperature dependence of the magnetic contribution of $\rho_m$ for resistivity of Ce$_2$PdGa$_{12}$ under several pressures.

Figure 3. $\rho_m$ at low temperature region in a linear scale. Dotted and broken arrows indicate the Neel temperature $T_N$ and $T_{shoulder}$ (see text).

Figure 4. Pressure dependence of $T_{CEF}$, $T_{shoulder}$ and $T_N$ for Ce$_2$PdGa$_{12}$. 
pressure-induced superconductor CeNiGe$_3$[11].

Figure 4 shows pressure dependence of the characteristic temperatures of $T_{CEF}$, $T_{shoulder}$ and $T_N$ for Ce$_2$PdGa$_{12}$ in a semi-logarithmic scale. Solid lines are eyeguide line. $T_{CEF}$ shows minimum around $T_{CEF}$ at $P_c \sim 7$GPa. $T_N$ is rapidly suppressed by pressure up to 3GPa. Between 2 and 6 GPa, $T_{shoulder}$ is appeared. $T_{shoulder}$ may reflect coherent Kondo scattering and/or magnetic scattering. In either case, $T_{shoulder}$ is disappeared at 8GPa. A more detailed study for magnetic properties under high pressure is required to assign $T_{shoulder}$.

4. Summary
We have synthesized clean single crystals of tetragonal Ce$_2$PdGa$_{12}$ and done the pressure study on the electrical resistivity. The electronic state of Ce$_2$PdGa$_{12}$ are tuned by pressure from the heavy-fermion antiferromagnetic state to the non-magnetic heavy-fermion state. We speculate that the non-magnetic ground state of Ce$_2$PdGa$_{12}$ might appear around $P_c \sim 7$GPa. In order to look for possible superconductivity around $P_c$ for Ce$_2$PdGa$_{12}$, advanced measurements in the lower temperature region are currently underway.

Acknowledgments
This work was supported by a Grant-Aid for Scientific Research on Innovative Areas ”Heavy Electrons” (No. A01-23102712) from The Ministry of Education, Culture, Sports, Science, and Technology, Japan.

References
[1] See e.g., Doniach S 1977 Valence Instability and Related Narrow Band Phenomena, ed. Parks R D (New York: Plenum)
[2] Mathur N D, Grosche F M, Julian S R, Walker I R, Freye D M, Haselwimmer R K W and Lonzarich G G 1998 Nature \textbf{394} 39.
[3] Assmus W, Herrmann M, Rauchschwalbe U, Riegel S, Lieke W, Spille H, Horn S, Weber G, Steglich F and Cordier G 1984 \textit{Phys. Rev. Lett.} \textbf{52} 469.
[4] Petrovic C, Pagliuso P G, Hundley M F, Movshovich R, Sarrao J L, Thompson J D, Fisk Z and Monthoux P 2001 \textit{J. Phys.: Condens. Matter} \textbf{13} L337.
[5] Hegger H, Petrovic C, Moshopoulou E G, Hundley M F, Sarrao J L, Fisk Z, and Thompson J D 2000 \textit{Phys. Rev. Lett.} \textbf{84} 4986.
[6] Chen G, Ohara S, Hedo M, Uwatoko Y, Saito K, Sorai M and Sakamoto I 2002 \textit{J. Phys. Soc. Jpn.} \textbf{71} 2836.
[7] Nicklas M, Sidorov V A, Borges H A, Pagliuso P G, Petrovic C, Fisk Z, Sarrao J L, and Thompson J D 2003 \textit{Phys. Rev. B} \textbf{67} 020506.
[8] Macaluso R T, Millican J N, Nakatsuji S, Lee S H, Carter B, Moreno N O, Fisk Z and Chan J Y 2005 \textit{Journal of Solid State Chemistry} \textbf{178} 3547.
[9] Cho J Y, Millican J N, Capan C, Sokolov D A, Molenovan M, Karki A M, Young D P, Aronson M C and Chan J Y 2008 \textit{Chem. Mater} \textbf{20} 6116.
[10] Thompson J D and Lawrence J L 1994 \textit{Handbook on the physics and Chemistry of Rare Earths} 19, ed. Gschneidner K A, Eyring L, Lander G H, Choppin C (North-Holland: Amsterdam) 383.
[11] Nakashima M, Tabata K, Thamizhavel A, Kobayashi T C, Hedo M, Uwatoko Y, Shimizu K, Settai R and Onuki Y 2004 \textit{J. Phys.: Condens. Matter} \textbf{16} L255.