Specific heat of Ce$_3$Pd$_{20}$Si$_6$ single crystal in magnetic fields

H Ono$^1$, T Nakano$^2$, N Takeda$^2$, G Ano$^1$, M Akatsu$^1$, Y Nemoto$^1$, T Goto$^1$ and H Kitazawa$^3$

$^1$ Graduate School of Science and Technology, Niigata University, Niigata, Japan
$^2$ Faculty of Engineering, Niigata University, Niigata, Japan
$^3$ National Institute for Materials Science, Tsukuba, Japan
E-mail: f09k001f@mail.cc.niigata-u.ac.jp

Abstract. We report the specific heat measurement of single crystalline Ce$_3$Pd$_{20}$Si$_6$ and the resultant magnetic phase diagram for the direction of $B$ // [100]. In zero field, we confirmed two successive phase transitions at $T = 0.24$ K and 0.4 K. The magnetic phase diagram clearly shows a phase boundary of paramagnetic phase I, quadrupole ordered phase II and II$'$. 

1. Introduction

The crystal structure of Ce$_3$Pd$_{20}$Si$_6$ is a Cr$_{23}$C$_6$-type cubic with a space group $Fm\bar{3}m$ (No. 225) [1]. Ce atoms occupy two different crystallographic sites; Ce atom at 4a-site is inside a cage of 12 Pd atoms and 6 X atoms, and forms a face-centered cubic lattice, the other Ce atom at 8c-site is surrounded by a cage of 16 Pd atoms, which forms a simple cubic lattice.

The magnetic properties of Ce$_3$Pd$_{20}$Si$_6$ at low temperatures are controversial. In an earlier investigation, Ce$_3$Pd$_{20}$Si$_6$ is a spin glass below $T = 0.15$ K [2], afterward, it was found that Ce$_3$Pd$_{20}$Si$_6$ shows two successive phase transitions at $T_L = 0.31$ K and $T_U = 0.5$ K [3]. The effect of $T_L$ and $T_U$ on magnetic field is different. $T_L$ shifts to lower temperature with increase magnetic field and vanishes above 1 T, whereas $T_U$ shifts to higher temperature and becomes unclear above 8 T. Very recently, Goto et al. measured the elastic constant on single crystal for three principal axes and constructed the magnetic phase diagram [4]. They found that Ce$_3$Pd$_{20}$Si$_6$ has three ordered phases II, II$'$, III and paramagnetic phase I (refer to Fig. 4 which will be described later). The phase III is antiferromagnetic and the phase II and II$'$ are identified as antiferro-quadrupole ordered phases. The phase boundary between phase-I and phase-II is strongly anisotropic in the magnetic field. In zero field, however, they found only one transition at 0.17 K. Mitamura et al. carried out magnetization measurements for principal axes using specimen cut from the same single crystal used in ultrasonic measurements [5]. The magnetic phase diagram obtained from magnetization measurements has two phase transitions at 0.23 K.
and 0.45 K. Slightly lower transition temperature than $T_L$ and $T_U$ is due to a strong sample dependence which is thought to be due to sample homogeneity and annealing conditions [6].

As mentioned above, the low temperature physical properties of Ce$_3$Pd$_{30}$Si$_6$ is still controversial even in the same single crystal. The purpose of the present study is to establish the magnetic phase diagram with $B \parallel [100]$ by specific heat measurement.

2. Experimental

Starting materials were 5N-Ce, 3N5-Pd and 10N-Si. They were mixed with an atomic ratio Ce : Pd : Si = 3 : 20 : 6, and melted into a button in an argon atmosphere using a mono-arc furnace. A single crystal of Ce$_3$Pd$_{30}$Si$_6$ was grown by a floating zone furnace equipped with double ellipsoidal mirrors. The specimen used in the present study was cut from the same single crystal used in ultrasound and magnetization measurements and annealed for 4 weeks at 1173 K. The specific heat was measured by an adiabatic heat pulse method with a dilution refrigerator.

3. Results and Discussions

Figure 1 shows the temperature dependence of the specific heat, $C(T)$, of Ce$_3$Pd$_{30}$Si$_6$ for $B \leq 1.0$ T. In zero field, we observed a sharp peak at $T = 0.24$ K and a shoulder at $T = 0.4$ K, which indicates antiferromagnetic and antiferro-quadrupole transition. Hereafter, we use a notation of $T_{II-III}$ and $T_{I-II}$ etc. as a transition temperature. The sharp peak at $T_{II-III}$ shifts to lower temperature with increasing magnetic field and vanishes at $B = 1$ T. On the other hand, $T_{I-II}$ shifts to higher temperature with increasing $B$. These behaviors are consistent with the magnetization measurement.

Figure 2 shows $C(T)$ for $1.0$ T $\leq B \leq 2.5$ T. It has to be noted that simple extrapolation of $C(T)$ at $B = 1.0$ T to $T = 0$ K does not go to zero. Since $C(T)$ goes to zero at $T = 0$ K, $C(T)$ at $B = 1.0$ T should decrease rapidly below 0.1 K. Although it is difficult to see a sign of decrease in $B = 1.0$ T data, a faint peak appears around 0.15 K above 1.2 T. The faint peak for $B \leq 1.8$ T is too tiny.
to conclude as a phase transition, but it becomes clear above 2.0 T and moves to higher
temperature and merges with a peak at $T_{I-II}$. This peak above $B = 2.0$ T just corresponds to the
transition of phase II to II’.

Figure 3 exhibits $C(T)$ above 2.5 T. The peak indicates the transition between phase II’ and I. $T_{I-II}$ decreases monotonically and vanishes at 4 T.

![Figure 2](image2)

Figure 2. (Color online) The temperature dependence of specific heat. The date from bottom to top show $B = 1.0, 1.2, 1.5, 1.8, 2.0, 2.1, 2.15, 2.2, 2.3$ and 2.5 T, which are shifted by 1 J / K mol-Ce, respectively. The arrow indicates the transition between phase II and II’.

![Figure 3](image3)

Figure 3. (Color online) The temperature dependence of specific heat of Ce$_3$Pd$_{20}$Si$_6$ for $B \geq 2.5$ T. This peak indicates the transition between phase I and II’.

We show in Figure 4 the magnetic phase diagram of Ce$_3$Pd$_{20}$Si$_6$ for the $B // [100]$ obtained from the present measurements. We plot a peak temperature in $C$ as a phase transition temperature. The transition temperature $T_{I-II}$ significantly increases with $B$ at low fields. This is a common feature of quadrupole transition. The phase boundary between paramagnetic phase I and quadrupole ordered phase II or II’ around $B = 2$ T clearly shows a cusp structure, which has not been observed in the magnetization and elastic constant measurements. This cusp structure would suggest that the order parameter of two quadrupole ordered phase have a competitive
effect each other.

Figure 4. (Color online) The magnetic phase diagram of Ce$_3$Pd$_{20}$Si$_6$ for $B // [100]$, determined from the present specific heat measurements. The phase I is paramagnetic phase. The phase II and II’ are antiferro-quadrupole ordered phase. The phase III is antiferromagnetic ordered phase.

4. Conclusion

We measured the specific heat of Ce$_3$Pd$_{20}$Si$_6$ and constructed the magnetic phase diagram with $B // [100]$. We confirmed two successive transition at $B = 0$ T. The phase diagram below 1T is in good accordance with the magnetization measurement.

We established clear phase boundary between I, II and II’ around $B = 2$ T for the first time. The faint peak around 0.15 K for 1.2 T $\leq B \leq$ 1.8 T is still ambiguous.

Acknowledgment

This work was supported by a Grand-Aid of Scientific Research on Innovative Area “Heavy Fermion” (No.20102004) of Ministry of Education, Culture, Sports, Science and Technology, Japan.

4. References

[1] Grivanov A V, Seropdgin Yu D and Bodak O I, (1994) J. Alloys. Comp. 204 L9
[2] Takeda N, Kitagawa J and Ishikawa M, (1995) J. Phys. Soc. Japan 64 387
[3] Strydom A M, Pikul A, Steglich F and Paschen S, (2006) J. Phys.: Conf. Ser. 51 239
[4] Goto T, Watanabe T, Tsuduku S, Kobayashi H, Nemoto Y, Yanagisawa T, Akatsu M, Ano G, Suzuki O, Takeda N, Dönni A and Kitazawa H, (2009) J. Phys. Soc. Japan 78 024716
[5] Mitamura H, Tayama T, Sakakibara T, Tuduku S, Ano G, Ishii I, Akatsu M, Nemoto Y, Goto T, Kikkawa A and Kitazawa H, (2010) J. Phys. Soc. Japan 79 0747102
[6] Shiobara D, Master Thesis (2009) Niigata University