Journal of Physics: Conference Series **592** (2015) 012022 doi:10.1088/1742-6596/592/1/012022

Single crystal growth and physical properties of $YbPd_2Si_2$

Y Matsumoto[1], K Hanya[1], Y Haga[2], Z Fisk[2,3], and S Ohara[1]

[1]Department of Engineering Physics, Electronics and Mechanics, Graduate school of Engineering, Nagoya Institute of Technology, Nagoya, Aichi 466-8555, Japan
[2]Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan
[3]Department of Physics and Astronomy, University of California, Irvine, California 92697, USA

E-mail: matsumoto.yuji@nitech.ac.jp

Abstract.

We report the heat capacity and magnetic properties of single crystals of $YbPd_2Si_2$ grown from Sn flux. $YbPd_2Si_2$ is a non magnetic heavy fermion compound with electronic specific heat coefficient γ= 95 mJ/mol K^2 and Pauli paramagnetic susceptibility χ_0 = 0.0115 emu/mol. The density of states of single crystal $YbPd_2Si_2$ is approximately half that of polycrystalline $YbPd_2Si_2$ while the strength of the Kondo effect in the single crystal is approximately twice that of poly crystal.

1. Introduction

In the study of quantum critical phenomena in highly correlated f electron systems, Yb compounds have been found to exhibit interesting quantum phenomena. β-$YbAlB_4$ exhibits heavy fermion superconductivity [1], YbT_2Zn_{20} (T=transition metal) exhibits heavy fermion behavior where the electronic specific heat coefficient γ of $YbCo_2Zn_{20}$ reaches about 8 J/mol K^2 [2], and ferromagnetic quantum fluctuations have been observed in $YbNi_4P_2$ [3].

The tetragonal $ThCr_2Si_2$-type structure series is one of the most studied group of compounds. Among the Yb member compounds, $YbRh_2Si_2$ exhibits anomalous quantum critical behavior which cannot be explained by the conventional Hertz-Millis-Moriya (MHH) theory [4, 5, 6, 7]. In the paramagnetic heavy fermion compounds $YbCu_2Si_2$ and $YbNi_2Ge_2$ [8, 9], pressure induced magnetic order takes place. And superconductivity occurs in $YbPd_2Ge_2$ [10].

$YbPd_2Si_2$ is paramagnetic heavy fermion compound with $ThCr_2Si_2$-type structure whose γ is approximately 200 mJ/mol K^2 [11]. The lattice volume V is 165.074 $Å^3$ [12]. On the other hand, $YbNi_2Si_2$ with the same electron count is an antiferromagnet with T_N = 2.3 K and V is 145.736$Å^3$ [13]. If $YbNi_2Si_2$ and $YbPd_2Si_2$ fit on the so-called Doniach phase diagram, then $YbPd_2Si_2$ will exhibit antiferromagnetic order under pressure. Actually, magnetic order has been observed above the critical pressure p_c = 1 GPa [14]. These pressure measurements have been performed on a polycrystalline sample with residual resistivity ratio (RRR) and residual resistivity of about 2 and 90 $\mu\Omega$cm, respectively. High quality single crystals are necessary for the study of quantum critical phenomena. However, there is no report of the single crystal

growth for $YbPd_2Si_2$. Therefore we have synthesized and performed magnetic property and specific heat measurements on $YbPd_2Si_2$ single crystal grown from Sn-flux.

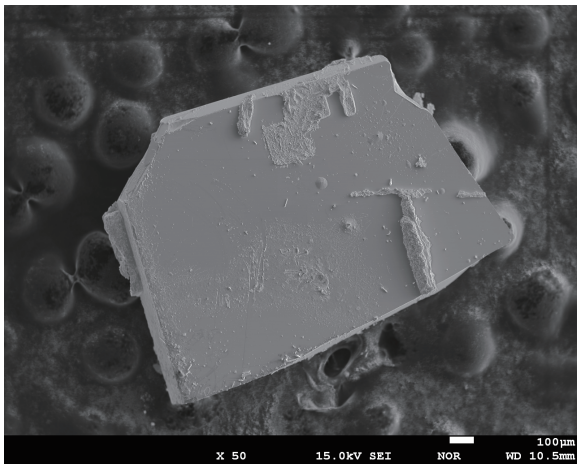

Figure 1. Scanning electron microscopic images of $YbPd_2Si_2$.

2. Experimental

The single crystal of $YbPd_2Si_2$ were grown in Sn-flux method. 3N (99.9 %) Yb, 3N Pd, 6N Si and 6N Sn were reacted in the starting composition of 1:2:2:50. These materials were placed in an alumina crucible and sealed in an evacuated quartz tube. The sealed tubes were heated to 1150 °C, soaked for 12 hours, then cooled down to 500 °C in 96 hours. The excess Sn was spun off in a centrifuge. Figure 1 shows the scanning electron microscopic images of $YbPd_2Si_2$.

The single phase of the tetragonal $ThCr_2Si_2$ structure was confirmed by the powder X-ray diffraction method. The crystal compositions and homogeneity were determined by using an electron micro-probe with wavelength dispersive spectrometers (electron probe micro analyzer(EPMA)-WDS; JEOL-8530F). We used the YbP_5O_{14}, Pd and Si as standard reference materials for EPMA. The chemical composition was determined to be Yb:Pd:Si = 1.00:1.83:2.08, indicating that the values are similar to the stoichiometric compositions of $YbPd_2Si_2$, although the ratio of Pd is slightly small.

The magnetic susceptibility was measured using a commercial superconducting quantum interference device magnetometer (Quantum Design). The specific heat measurement was carried out by the relaxation method using a commercial physical property measurement system (PPMS; Quantum Design).

3. Results and discussion

Figure 2 shows the magnetic susceptibility as a function of temperature at 1 T with magnetic field applied parallel to the [001] direction. With decreasing temperature, the susceptibility increases to a broad maximum. In heavy fermion compounds such as $CeRu_2Si_2$ [15], such a broad maximum have been observed. Here the temperature of the maximum of susceptibility $T\chi_{max}$ is 41 K. The $T\chi_{max}$ of our single crystal is about two times that of the polycrystalline samples of the previous study. In the low temperature region, the susceptibility slightly increases with decreasing temperature, probably due to magnetic impurities. The maximum of susceptibility around $T\chi_{max}$ is wider than that of poly crystal [16]. It would be caused by the impurity phases. If we neglect the magnetic impurity, we estimated χ(T=0) = 0.0115 emu/mol, a large Pauli paramagnetic value.

Figure 3 shows the temperature dependence of the reciprocal magnetic susceptibility. Above 150 K, the temperature dependence of the inverse magnetic susceptibility can be fit by a

Curie-Weiss law with effective moment $\mu_{\rm eff}$ and Weiss temperature $\theta_{\rm p}$ of 4.54 $\mu_{\rm B}$ and -123 K, respectively, the effective moment being very close to the Hund's Rule value expected for Yb^{+3} $\mu_{\rm eff} = 4.53$ $\mu_{\rm B}$.

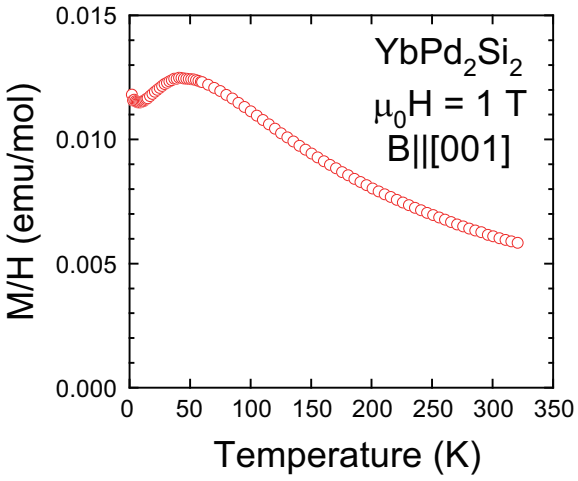

Figure 2. Temperature dependence of the magnetic susceptibility at 1 T for a magnetic field along [001] direction.

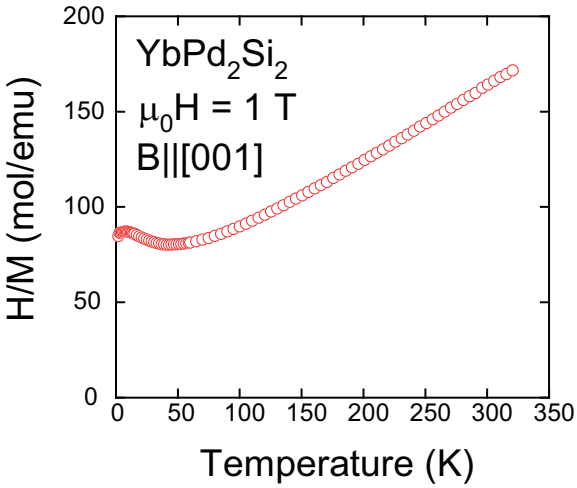

Figure 3. Temperature dependence of the reciprocal magnetic susceptibility at 1 T for a magnetic field along [001] direction.

Figure 4 shows the temperature dependence of the specific heat C. The inset in Fig. 4 shows C/T versus T^2. Using the expression $C/T = \gamma + \beta\ T^2$, we estimate the value of the specific heat coefficient $\gamma = 95$ mJ/mol K^2 and $\beta = 0.555$ mJ/mol K^4. This γ of our single crystal is approximately half that reported for the polycrystal.

The physical properties of single crystal $YbPd_2Si_2$ are similar to that of polycrystalline, indicating that single crystal $YbPd_2Si_2$ is a paramagnetic heavy fermion. However the γ, $T\chi_{max}$ and χ_0 differ between them. We summarize the physical properties of single crystal and polycrystal$YbPd_2Si_2$ in table 1. χ_0 and γ are proportional to the density of states (DOS). χ_0 and γ of single crystal are about half those of polycrystal, indicating a DOS of single crystal about half that of polycrystal. In heavy fermion systems, $T\chi_{max}$ is proportional to the Kondo temperature $T_{\rm K}$ [15], indicating that $T_{\rm K}$ of single crystal is about twice larger than that of poly crystal. Moreover, the electronic specific coefficient γ is roughly in inverse proportional to $T_{\rm K}$ [17]. γ is approximately half that of poly crystal. The difference of physical properties between the single crystal and polycrystal could be caused by the change of strength of Kondo effect. The strength of Kondo effect is changed by the electron or hole doping, the change of lattice volume etc. The defect at the Pd site can possibly change the number of electron and lattice volume. The difference of physical properties between single crystal and polycrystal could be caused by the defect at the Pd site.

4. Conclusion

We have synthesized single crystal $YbPd_2Si_2$ from Sn flux. The crystal structure was confirmed by powder X-ray diffraction and chemical composition was determined to be Yb:Pd:Si = 1.00:1.83:2.08. We have measured the specific heat and susceptibility of $YbPd_2Si_2$. It was found that single crystal $YbPd_2Si_2$ is a non magnetic heavy fermion compound with the $\gamma = 95$ mJ/mol K^2 and $\chi_0 = 0.0115$ emu/mol.

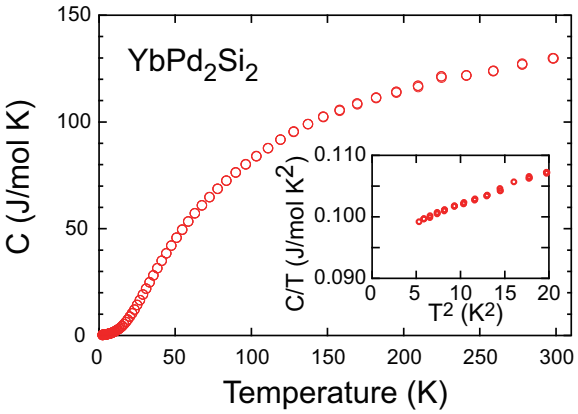

Figure 4. Temperature dependence of specific heat. The inset shows the C/T vs. T^2.

Table 1. χ_0, $T\chi_{max}$ and γ of poly crystal and single crystal .

sample	χ_0 (emu/mol)	$T\chi_{max}$ (K)	γ (mJ/mol K^2)
poly crystal (arc melt)	0.018 [16]	25 [16]	203 [11]
single crystal (Sn flux (this work))	0.0115	41	95

Acknowledgments
We thank A. Iwasaka for technical support of EPMA measurements. We thank M. Taki and Y. Minamiguchi for technical support. This work was supported by a Grant-in-Aid for Scientific Research Young Scientists B (No 24740248) from the Ministry of Education, Culture, Sports, Science and Technology (MEXT) and Japan Society of the Promotion of Science (JSPS).

References
[1] Nakatsuji S, Kuga K, Machida Y, Tayama T, Sakakibara T, Karaki Y, Ishimoto H, Yonezawa S, Maeno Y, Pearson E, Lonzarich G G, Balicas L, Lee H, and Fisk Z, 2008*Nature Phys.* **4** 603.
[2] Torikachavili M S, Jia S, Mun E D, Hannahs S T, Black R C, Neils W K, Martien D, Bud'ko S L, and Canfield P C, 2007 *Proc. Natl. Acad. Sci* **104** 9960.
[3] Krellner C, Lausberg S, Steppke A, Brando M, Pedrero L, Pfau H, Tence S, Rosner H, Steglich F, and Geibel C, 2012 *New J. Phys.* **13** 103014.
[4] Custers J, Gegenwart P, Wilhelm H, Neumaier K, Tokiwa Y, Trovarelli O, Geibel C, Fseglich F, Pepin C, and Coleman, 2002 *Nature* **424** 524.
[5] Hertz, J A, 1976, *Phys. Rev. B* **14** 1165.
[6] Millis A J, 1993 *Phys. Rev. B* **48** 7183.
[7] Moriya T, and Takimoto T, 1995 *J. Phys. Soc. Jpn* **64** 960.
[8] Knebel G, Braithwaite D, Lapertoto, Canfiled P C, Flouquet J, 2001 *J. Phys.:Condens. Matter.* **13** 10935.
[9] Tateiwa N, Matsuda T D, Haga Y, and Fisk Z, 2014 *Phys. Rev. B* **89** 035127.
[10] Hull G W, Wernick J H, Geballe T H, Waszczak J V, Bernardini J E, 1981 *Phys. Rev. B* **24** 6715.
[11] Dhar S K, Sampathkumaran E V, and Vijayaraghavan R, 1987 *Solid State Commun.* **61** 479.
[12] Yamaoka H, JArrige I, Tsuji N, Lin J-F Hiraoka N, Ishii H, and Tsuei K-D, 2010 *Phys. Rev. B* **82** 035111.
[13] Andre G, Bonville P, Bouree F, Bombik A, Kolenda M, Oles A, Pacyna A, Sikora W, and Szytula A, 1995 *J. Alloys Comp.* **224** 253.
[14] Nakano T, Hedo M, Uwatoko Y, and Sampathkumaran E V, 2004 *Solid Sate Commun.* **132** 325.
[15] Haen P, Flouquet J, Lapierre F, Lejay P, and Remenyi G, 1987 *J. Low Temp. Phys.* **67** 391.
[16] Bonville P, Hammann J, Hodges J A, Imbert P, Jehanno G, Besnus M J, and Meyer A, 1991 *Z. Phys. B Condensed Matter* **82** 267.
[17] Onuki Y, Settai R, Sugiyama K, Takeuchi T, Kobayashi T C, Haga Y, and Yamamoto E, 2004 *J. Phys. Soc. Jpn.* **73** 769.