Journal of Alloys and Compounds, 213/214 (1994) 387–390
JALCOM 4077

Effect of pressure on the electrical resistivity of URu_2Si_2

Tomoko Kagayama[a], Gendo Oomi[a], Keiko Iki[a], Nobuo Mōri[b], Yoshichika Ōnuki[c] and Takemi Komatsubara[d]

[a]*Department of Physics, Faculty of General Education, Kumamoto University, Kumamoto 860 (Japan)*
[b]*Institute for Solid State Physics, The University of Tokyo, Tokyo 106 (Japan)*
[c]*Institute of Materials Science, University of Tsukuba, Ibaraki 305 (Japan)*
[d]*Department of Physics, Tohoku University, Sendai 980 (Japan)*

Abstract

The electrical resistivity $\rho(T)$ of the concentrated Kondo compound URu_2Si_2 has been measured at high pressure up to 80 kbar in order to examine the stability of the electronic state at high pressure. The pressure dependence of volume is also observed by X-ray diffraction. The Grüneisen parameters for the Néel temperature and the Kondo temperature are estimated on the basis of the results in the present work. The present analysis emphasizes the itinerant character of 5f electrons of URu_2Si_2.

1. Introduction

The physical properties of the concentrated Kondo (CK) compounds are well known to be characterized by two temperatures T_{RKKY} and T_K, where T_{RKKY} indicates the intensity of the indirect Ruderman–Kittel–Kasuya–Yoshida (RKKY) interaction and T_K is the Kondo temperature. Since these two interactions compete with each other, a variety of physical properties have been observed in the CK systems including U, Ce and Yb elements [1, 2]. If T_{RKKY} is larger than T_K, a magnetic ordered state would be stabilized in the system and vice versa. T_K and T_{RKKY} are expressed as follows [1]:

$$T_K \propto \exp\left(-\frac{1}{|JN(0)|}\right) \tag{1}$$

$$T_{RKKY} \propto \frac{J^2}{W} \tag{2}$$

where J is the Kondo coupling constant, $N(0)$ the density of states at the Fermi level and W the band width of conduction electrons. $|J|$ is related to the magnitude of hybridization V_{sf} and the energy difference $\Delta\epsilon$ between 4f level and Fermi level as $|J| \propto V_{sf}^2/|\Delta\epsilon|$. Since V_{sf} increases and $|\Delta\epsilon|$ decreases with increasing pressure, $|J|$ is expected to increase on application of pressure. For this reason, both T_K and T_{RKKY} change drastically at high pressure to induce a lot of interesting features in CK compounds.

The compound URu_2Si_2 has been studied extensively because it exhibits two electronic phase transitions at low temperatures: the one is superconductivity near 1.5 K (*i.e.* T_c) and the other the antiferromagnetic order at 17.5 K (*i.e.* T_N) [3, 4]. It is also characterized as a heavy fermion (HF) having a large electronic specific heat coefficient $\gamma \approx 65$ mJ mol^{-1} K^{-2} and a resistance maximum near 75 K (*i.e.* T_{max}). We have emphasized until now that the value of $|JN(0)|$ is a very important factor to determine the physical properties of HF compounds including elemental Ce [5], which is expected from eqns. (1) and (2). $JN(0)$ is usually estimated from the pressure dependence of T_{max} [6, 7], the value of the coefficient of T^2 [5] in the electrical resistivity $\rho(T)$ or ρ *vs.* log T plot.

In the present work we made an attempt to measure the electrical resistivity $\rho(T)$ under high pressure up to 80 kbar to clarify the electronic state of the HF compound URu_2Si_2 at high pressure by an assessment of the value of the Grüneisen parameter and $|JN(0)|$. The pressure dependence of volume is also measured at room temperature to examine the crystal structure change induced by pressure and the compressibility. The results will be discussed on the basis of the phenomenological treatment developed by Kagayama and Oomi [5] and then the value of $|JN(0)|$ will be obtained.

2. Experimental procedure

2.1. Sample and electrical resistance measurement at high pressure

The polycrystalline URu_2Si_2 sample was prepared by arc melting weighed amounts of pure U, Ru (99.999%)

0925-8388/94/$07.00 © 1994 Elsevier Science S.A. All rights reserved
SSDI 0925-8388(94)04077-H

and Si (99.999%) followed by annealing at 900 °C for one week.

Electrical resistance measurements were carried out in the temperature range between 2 K and 300 K by using a standard four-probe method in which four gold leads (diameters 20 and 50 μm) were attached to the sample using gold paste or spot welding. Hydrostatic pressure below 20 kbar was generated by using a standard Cu–Be piston–cylinder device and above 20 kbar by using a cubic-anvil press. A 1:1 mixture of transformer oil and kerosene and a mixture of Fluorinert, FC 70 and FC 77 were used as pressure transmitting media below 20 kbar and above 20 kbar respectively. The pressure was changed only at room temperature in order to minimize the internal strain in the sample and kept constant within $\pm 1\%$ throughout each measurement. The details of the high pressure system were reported elsewhere [8, 9].

2.2. X-ray diffraction study at high pressure

Hydrostatic pressure for X-ray diffraction was generated by using Bridgman-type tungsten carbide opposed anvils having a face 3 mm in diameter. The powdered sample and NaCl were placed in a 0.3 mm hole at the centre of a disk-shaped beryllium pressure vessel 0.5 mm in thickness [10]. A 4:1 mixture of methanol and ethanol was used as a pressure transmitting medium. The pressure was determined by using Decker's equation of state for NaCl [11].

3. Results

3.1. Temperature dependent electrical resistivity at high pressures

Figure 1 shows the electrical resistivity as a function of temperature in the form of $\Delta\rho = \rho - \rho_0$ at various pressures above 5 kbar, where ρ is the observed resistivity and ρ_0 the residual resistivity. ρ_0 is found to be almost independent of pressure having a value of 15 $\mu\Omega$ cm. $\Delta\rho$ at 5 kbar increases with increasing temperature, exhibits a hump near 18 K and has a broad maximum centred near 87 K (T_{max}). $\Delta\rho$ at 300 K is also almost independent of pressure. T_{max} increases with increasing pressure but the peak in the $\Delta\rho(T)$ curve becomes less clear at high pressure. It should be noted that the pressure derivative $\partial\rho/\partial P$ of ρ is very large near 80 K while the values of $\partial\rho/\partial P$ are almost 0 at 0 K and 300 K. Such behaviour is in sharp contrast with that of Ce compounds [6]. The pressure dependence of T_{max} is shown in Fig. 2. T_{max} increases with pressure having a rate $\partial T_{max}/\partial P = 1.3$ K kbar^{-1}, which is in good agreement with the previous lower pressure result [12] but smaller than those of Ce HF compounds, $\partial T_{max}/\partial P \approx 3$–4 K kbar^{-1} [6].

Figure 3 shows the Néel temperature T_N as a function of pressure. T_N increases with increasing pressure having an initial pressure coefficient $\partial T_N/\partial P \approx 0.16$ K kbar^{-1}, which is in good agreement with the previous result [13]. However, the increasing rate of T_N becomes sluggish

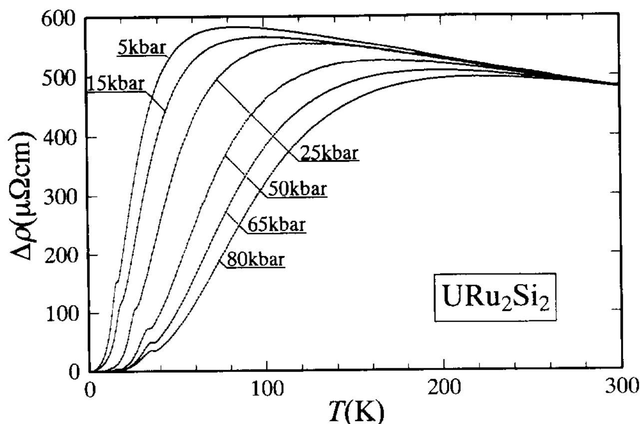

Fig. 1. The temperature-dependent electrical resistivity $\Delta\rho \equiv \rho - \rho_0$ at various pressures above 5 kbar. ρ is the observed resistivity and ρ_0 the residual resistivity.

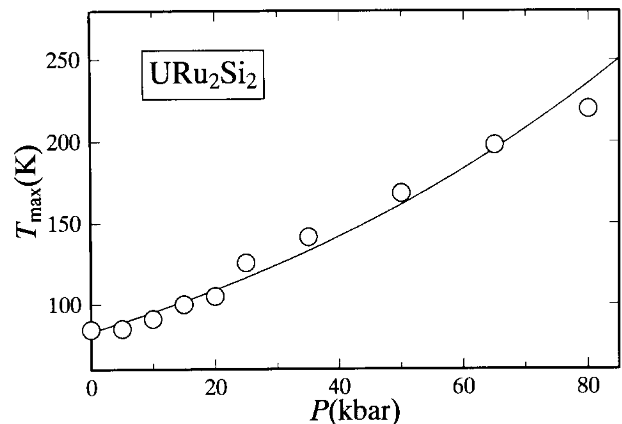

Fig. 2. Pressure dependence of the resistivity-maximum temperature T_{max}: ——, the fitting result mentioned in section 4.

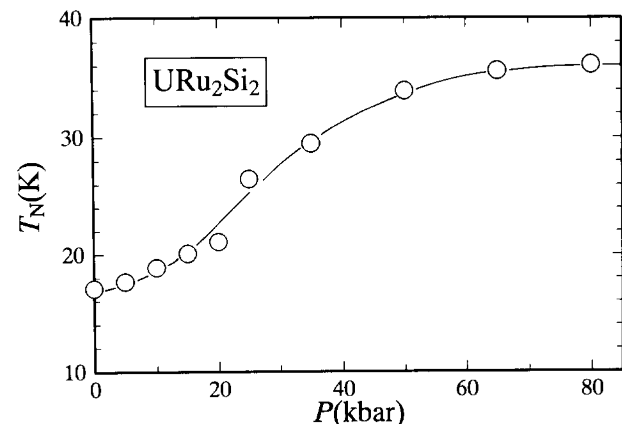

Fig. 3. The Néel temperature T_N as a function of pressure.

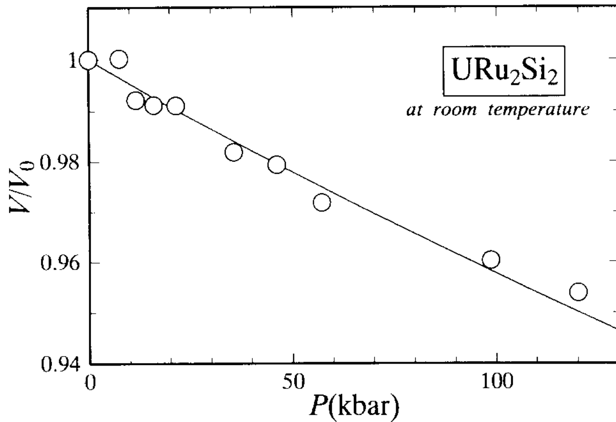

Fig. 4. The relative change V/V_0 in volume as a function of pressure at room temperature.

above 50 kbar to give rise to a broad maximum in the T_N *vs.* P curve around 80 kbar ($T_N \approx 36$ K), which may be caused by a competition between the RKKY interaction and the Kondo coupling [1, 14].

3.2. Compression curve of URu_2Si_2 at room temperature

The relative change V/V_0 in volume is illustrated in Fig. 4 as a function of pressure at room temperature. Since no new diffraction lines are observed up to 120 kbar, the $ThCr_2Si_2$-type crystal structure is stable in the present pressure range at room temperature. The volume is found to decrease smoothly with increasing pressure without any discontinuity. The bulk modulus $B = -V\,\partial P/\partial V$ and its pressure derivative are estimated by a least-squares fit to the following Murnaghan equation of state:

$$P = \frac{B_0}{B_0'}\left[\left(\frac{V}{V_0}\right)^{-B_0'} - 1\right] \tag{3}$$

where V and V_0 are the volume at respectively high and ambient pressure. The result of the least-squares fit is shown by the full curve in Fig. 4. The values B_0 and B_0' are obtained to be 2150 ± 40 kbar and 3.3 ± 0.2 respectively, which are slightly smaller than those obtained by Luo *et al.* [15]. It is found that B_0 is larger than those of Ce HF compounds [6, 10], which are as much as 1000 kbar, *i.e.* the U compound is harder than the Ce compound. The small derivative of T_{max} compared with that of the Ce compound may be partly a result of this fact.

4. Discussion

In this section we analyse quantitatively the pressure (or volume) dependence of T_K and T_{RKKY}. T_{max} is roughly proportional to T_K [7] and T_K is described as in eqn. (1). Thus the increase in T_{max} is explained as the increase in $|JN(0)|$. When the Grüneisen parameter for $|JN(0)|$ is denoted by q, $q \equiv -\partial \ln |JN(0)|/\partial \ln V$, the Grüneisen parameter Γ_K for T_K is written as [5]

$$\Gamma_K \equiv -\frac{\partial \ln T_K}{\partial \ln V} = -\frac{\partial \ln T_{max}}{\partial \ln V} = \frac{q}{|JN(0)|} \tag{4}$$

From eqn. (4), we derive the following equation:

$$\ln\left[\frac{T_{max}(P)}{T_{max}(0)}\right] = -\Gamma_K \ln\left(\frac{V}{V_0}\right) \tag{5}$$

To obtain the value of Γ_K we made an attempt to fit the present data to eqn. (5).

In Fig. 5 the value of $\ln[T_{max}(P)/T_{max}(0)]$ is plotted as a function of $-\ln(V/V_0)$, where the result in section 3.2 used as the relative change V/V_0 in the volume, and the fitting result is indicated as straight full line. From the slope of the line we obtained $\Gamma_K = 30$, which is about one-half of those for Ce compounds such as $CeInCu_2$ or $CeCu_6$ [16].

Next we estimate the Grüneisen parameter Γ_{RKKY} for T_{RKKY} in the same way as mentioned above. Since $T_{RKKY} \propto T_N$ the following equation is derived:

$$\ln\left[\frac{T_N(P)}{T_N(0)}\right] = -\Gamma_{RKKY} \ln\left(\frac{V}{V_0}\right) \tag{6}$$

Figure 6 shows the plot of $\ln[T_N(P)/T_N(0)]$ as a function of $-\ln(V/V_0)$. Because T_N saturates at high pressure owing to the competition between the RKKY interaction and Kondo effect, we used only the data below 20 kbar. Γ_{RKKY} is obtained to be 24. By using eqn. (2) and the relation $N(0) \propto 1/W$, we obtain $T_N \propto T_{RKKY} \propto |JN(0)|^2$. Then Γ_{RKKY} is written as $\Gamma_{RKKY} = 2q$. From the above estimation, the value of q is 12. Although q is usually taken to be around 6 [17], we obtained twice this value in the present es-

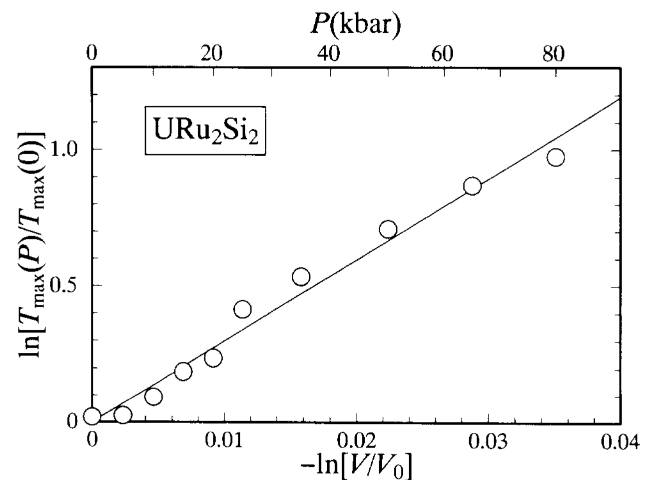

Fig. 5. The value of $\ln[T_{max}(P)/T_{max}(0)]$ as a function of $-\ln(V/V_0)$.

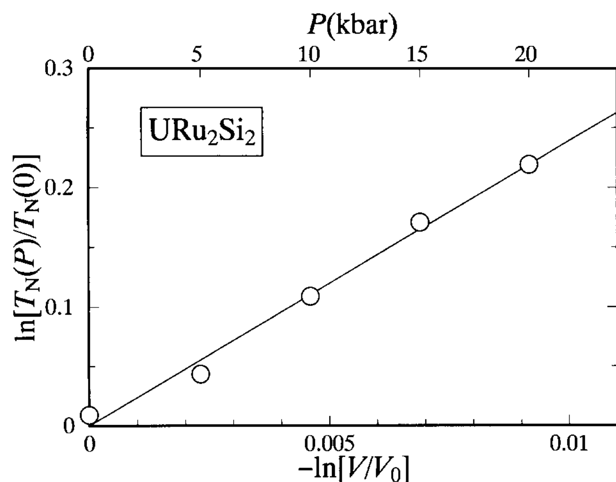

Fig. 6. The value of $\ln[T_N(P)/T_N(0)]$ as a function of $-\ln(V/V_0)$.

timation. From the value of Γ_K estimated in the foregoing paragraph, $|JN(0)|_0$ is obtained to be 0.4, which is larger than those of Ce HF compounds [5, 7, 16]. This implies the itinerant character of 5f electrons in U compounds.

5. Conclusion

We measured the temperature-dependent electrical resistivity $\rho(T)$ of the HF compound URu_2Si_2 up to 80 kbar. The volume is also measured as a function of pressure at room temperature. The main results are summarized as follows.

(1) The crystal structure of $ThCr_2Si_2$ type is stable at least up to 120 kbar at room temperature. The bulk modulus is 2150 kbar.

(2) The resistance-maximum temperature T_{max} increases with increasing pressure having an initial rate $\partial T_{max}/\partial P = 1.3$ K kbar^{-1}.

(3) The Néel temperature T_N increases with pressure but exhibits saturation above 50 kbar.

(4) The Grüneisen parameters for T_N and T_{max} are 24 and 30 respectively. From these values we estimated $|JN(0)|_0 \approx 0.4$. The magnitude of $|JN(0)|_0 \approx 0.4$ implies the itinerant character of the 5f electrons in URu_2Si_2.

References

1 N.B. Brandt and V.V. Moshchalkov, *Adv. Phys., 33* (1984) 373, and references cited therein.
2 J.D. Thompson, in L.C. Gupta and M.S. Multani (eds.), *Frontiers in Solid State Sciences,* Vol. 2, World Scientific, Singapore, 1992, p. 107.
3 T.T. Palstra, A.A. Menovsky, J. van den Berg, A.J. Dirkmaat, P.H. Kes, G.J. Nieuwenhuys and J.A. Mydosh, *Phys. Rev. Lett., 55* (1985) 2727.
4 M.B. Maple, J.W. Chen, Y. Dalichaouh, T. Kohara, C. Rossel, M.S. Torikachvili, M.W. McElfresh and J.D. Thompson, *Phys. Rev. Lett., 56* (1986) 185.
5 T. Kagayama and G. Oomi, *Transport and Thermal Properties of f-electron Systems, Hiroshima, 1992,* Plenum, New York, 1993, p. 155.
6 T. Kagayama and G. Oomi, *Jpn. J. Appl. Phys., 32* (1993) 318.
7 T. Kagayama, G. Oomi, H. Takahashi, M. Mōri, Y. Ōnuki and T. Komatsubara, *Phys. Rev. B, 44* (1991) 7690.
8 G. Oomi, T. Kagayama and Y. Uwatoko, *Jpn. J. Appl. Phys., 32* (1993) 349.
9 N. Mōri and H. Takahashi, *Pressure Eng. (J. High Pressure Inst. Jpn.), 28* (1990) 124 (in Japanese).
10 T. Kagayama, K. Suenaga, G. Oomi, Y. Ōnuki and T. Komatsubara, *J. Magn. Magn. Mater., 90–91* (1990) 451.
11 D.L. Decker, *J. Appl. Phys., 42* (1971) 3239.
12 M.W. McElfresh, J.D. Thompson, J.O. Willis and M.B. Maple, *Phys. Rev. B, 35* (1987) 43.
13 M.W. McElfresh, J.D. Thompson, J.O. Willis, M.B. Maple, T. Kohara and M.S. Torikachvili, *Phys. Rev. B, 35* (1987) 43.
14 S. Doniach, in R.D. Parks (ed.), *Valence Instabilities and Related Narrow-band Phenomena,* Plenum, New York, 1977, p. 169.
15 H. Luo, S. Dabos, U. Benedict and J.C. Spirlet, *J. Less-Common Met., 142* (1988) L23.
16 T. Kagayama, G. Oomi, H. Takahashi, N. Mōri, Y. Ōnuki and T. Komatsubara, *J. Alloys Comp., 181* (1992) 185.
17 M. Lavagna, C. Lacroix and M. Cyrot, *J. Phys. F, 13* (1983) 1007.