High pressure study on uranium heavy fermion compounds with antiferromagnetic ground state

N. Tateiwa\textsuperscript{1}, S. Ikeda\textsuperscript{1}, Y. Haga\textsuperscript{1}, T. D. Matsuda\textsuperscript{1}, M. Nakashima\textsuperscript{2}, D. Aoki\textsuperscript{3}, R. Settai\textsuperscript{4}, Y. ¯Onuki\textsuperscript{1,4}

\textsuperscript{1}A. S. R. C, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan
\textsuperscript{2}Faculty of Science, Shinshu University, Matsumoto, Nagano 390-8621, Japan
\textsuperscript{3}Institute for Nanoscience and Cryogenics, CEA/Grenoble, Grenoble 38054, France
\textsuperscript{4}Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan
E-mail: tateiwa.naoyuki@jaea.go.jp

Abstract. We have studied the high pressure effect on the antiferromagnetic ground states of uranium heavy fermion compounds. In this paper we show our high pressure studies on U\textsubscript{2}Zn\textsubscript{17} by the electrical resistivity measurements. In U\textsubscript{2}Zn\textsubscript{17}, the Néel temperature $T_N$ in U\textsubscript{2}Zn\textsubscript{17} decreases slightly from 9.66 K at 1 bar to 9.33 K at 2.5 GPa, becomes almost pressure-independent up to 4.7 GPa, and starts to increase at the higher pressure region. We discuss the pressure dependences of the coefficient of $T^2$ term in the electrical resistivity $A$, an antiferromagnetic gap $\Delta$ and a characteristic temperature $T_{\rho_{\text{max}}}$. It is found that the effect of pressure on the electronic states in U\textsubscript{2}Zn\textsubscript{17} is weak compared with the other heavy fermion compounds. The experimental results on UCu\textsubscript{5} is also discussed briefly.

1. Introduction

Application of pressure is an useful experimental method for controlling the magnetic interaction and hybridization between $f$ electrons and conduction electrons in uranium and cerium compounds, where the RKKY interaction and the Kondo effect compete with each other. As pressure is applied to compounds with magnetic orderings, the magnetic ordering temperature $T_{\text{mag}}$ of some compounds decreases and becomes zero at a critical pressure $P_c$: $T_{\text{mag}} \rightarrow 0$ at $P_c \rightarrow 0$, where the pressure-induced superconductivity or the non-Fermi liquid behavior appear\cite{1,2}. We have studied the high pressure effect on the antiferromagnetic ground state in typical uranium heavy fermion compounds UCu\textsubscript{5}, U\textsubscript{2}Zn\textsubscript{17}, and UCd\textsubscript{11} where the values of the linear specific heat coefficient $\gamma$ are far larger than 100 mJ/K\textsuperscript{2}·mol\textsubscript{U}. In this paper, we show our experimental results mainly on U\textsubscript{2}Zn\textsubscript{17}.

U\textsubscript{2}Zn\textsubscript{17} with the rhombohedral Th\textsubscript{2}Zn\textsubscript{17}-type structure (space group $R\bar{3}m$)\cite{3,4} shows an antiferromagnetic ordering at a Néel temperature $T_N = 9.7$ K at ambient pressure. The ordered magnetic moment $\mu_{\text{ord}}$ is 0.8 $\mu_B$/U\cite{5}. The specific heat coefficient $C/T$ shows a large value of about 500 mJ/K\textsuperscript{2}·mol\textsubscript{U} above $T_N$ but is reduced to about $\gamma = 200$ mJ/K\textsuperscript{2}·mol in $T \ll T_N$ \cite{3}. These results reveal the heavy fermion nature of an itinerant-$5f$ electronic state in U\textsubscript{2}Zn\textsubscript{17}. In this study, we have performed the electrical resistivity measurement under high pressures up to 9 GPa using a diamond anvil cell.
2. **Experiment**

A single crystal sample of U$_2$Zn$_{17}$ was grown by the Bridgman method with a W crucible sealed in high vacuum. The details of the sample preparation will be published in elsewhere. The value of residual resistivity ratio (RRR = $\rho_{RT}/\rho_0$) for a sample used in the present study is about 70, where $\rho_{RT}$ and $\rho_0$ are the resistivity at room temperature and the residual resistivity, respectively, indicating a comparably high quality of the present sample. For the electrical resistivity measurement, four gold-wires with 10 $\mu$m in diameter were bonded to a small sample with the dimensions $180 \times 50 \times 20 \mu m^3$ using a silver paste. We used a diamond anvil cell with the Dunstan and Spain-type [6, 7]. The sample and small ruby chips were clamped in a sample hole with 400 $\mu$m in diameter of a stainless steel gasket in DAC. The culet-size of diamonds is 800 $\mu$m. The noble gas argon (Ar) was used as a pressure-transmitting medium. It is well known that argon provides a good hydrostatic pressure up to 10 GPa [8]. The pressure was determined by the ruby fluorescence method at room temperature and 4.2 K [9]. A difference of pressures between at 4.2 K and 300 K is less than 5 %. The pressure determined at 4.2 K will be shown in following. The electrical resistivity under high pressure was measured by the dc 4-probe method using the $^4$He cryostat.

3. **Experimental results and discussions**

Figure 1 (a) shows the logarithmic temperature dependence of the electrical resistivity $\rho$ in the current parallel to the [1120] direction under high pressures. The inset of the Fig. 1 shows the resistivity at 1 bar. We show the low-temperature part of the electrical resistivity in Fig. 1 (b), where the Néel temperature $T_N$ is shown by an arrow. With decreasing temperature, the resistivity $\rho$ at 1 bar increases with a broad maximum at $T_{\rho_{\text{max}}} = 18.7$ K and shows a sharp kink at the Néel temperature $T_N = 9.65$ K. These are consistent with the previous study [3]. With increasing pressure, the Néel temperature $T_N$ decreases slightly from 9.66 K at 1 bar to 9.33 K at 2.54 GPa, stays almost constant up to 4.7 GPa, and then starts to increase at the higher pressure region. The value of $T_N$ is 12.2 K at 8.7 GPa. We show the pressure dependence of $T_N$ of U$_2$Zn$_{17}$ in Fig. 2 (a). A characteristic feature in U$_2$Zn$_{17}$ is that the sign of $dT_N/dT$ is

![Figure 1](image_url)
changed around 5 GPa. The previous study revealed that the Néel temperature $T_N$ shows a small pressure dependence up to 1.7 GPa, which is consistent with the present study [10, 11]. In the Fig. 2(a), we also plot the pressure dependence of $T_N$ in UCu$_5$ that orders antiferromagnetically at $T_N = 15.9$ K at ambient pressure [12]. The Néel temperature $T_N$ increases with increasing pressure simply up to 8 GPa [13].

The electrical resistivity below $T_N$ in U$_2$Zn$_{17}$ was analyzed by the antiferromagnetic magnon model described as

$$\rho = \rho_0 + AT^2 + BT(1 + \frac{T}{\Delta})\exp\left(-\frac{T}{\Delta}\right),$$

where the third term corresponds to the contribution from the electron scattering by the antiferromagnetic magnon with an energy gap $\Delta$ [14, 15]. We show a fit of the resistivity data by solid lines in Fig. 1 (b). Fig. 2 (b) shows the pressure dependences of obtained parameters $A$ and $\Delta$. The $A$ value decreases simply with increasing pressure, from 0.35 $\mu$Ω·cm/K$^2$ at 1 bar to 0.14 $\mu$Ω·cm/K$^2$. The corresponding $\gamma$ values are estimated as 190 and 120 mJ/K$^2$·mol at 1 bar and 8.7 GPa, respectively, using the Kadowaki-Woods relation ($A/\gamma^2 = 1.0 \times 10^{-5}$) [16]. The estimated $\gamma$ value at 1 bar is consistent with the observed value of about $\sim$ 200 mJ/K$^2$·molU [3]. The present result suggests that the $\gamma$ value decreases with increasing pressure. The value of $\Delta$ increases monotonously from 19 K at 1 bar to 33 K at 8.7 GPa.

The pressure dependence of $T_{p\max}$ is shown in the Fig. 2(c). The characteristic temperature $T_{p\max}$ increases linearly within increasing pressure shown as a solid straight line in the figure. The pressure derivative of $\partial T_{p\max}/\partial P$ is 1.0 K/GPa. $T_{p\max}$ corresponds to the characteristic temperature $T_0$ of the electronic state in U$_2$Zn$_{17}$. The Grüneisen parameter $\Gamma_{T_0}$ for $T_0$ is written as follows:

$$\Gamma_{T_0} = -\frac{\partial \ln T_0}{\partial \ln V} = B \frac{1}{T_0} \frac{\partial T_0}{\partial P},$$

where $B$ is the bulk modulus, estimated to be 83 GPa at 297 K for U$_2$Zn$_{17}$ from the ultrasound measurement [17]. The Grüneisen parameter $\Gamma_{T_0}$ is estimated as 4.6 in U$_2$Zn$_{17}$. This value is small, compared with heavy fermion compounds where the Grüneisen parameter is usually about one or two orders of magnitude larger than those in

![Figure 2](image-url)
ordinary metals [11, 18, 19].

The pressure effect on the antiferromagnetic state in U$_2$Zn$_{17}$ is discussed in comparison with cerium antiferromagnetic compounds CeIn$_3$ and CeRhIn$_5$ whose bulk moduli of 67 and 78, respectively, are close to that of U$_2$Zn$_{17}$ [20, 21]. Both CeIn$_3$ and CeRhIn$_5$ show the antiferromagnetic transition at $T_N$ =10 K and 3.8 K, respectively, at ambient pressure. Under high pressure, the Néel temperature $T_N$ becomes 0 K at the critical pressures $P_c$ of 2.5 and 2.1 GPa, respectively, and the pressure-induced superconductivity appears around $P_c$ [22, 23]. The magnetic-non magnetic transition takes place below 3 GPa in both cerium compounds whose compressibility $\beta$ (= 1/$B$) is similar to that of U$_2$Zn$_{17}$. On the other hand, the antiferromagnetic ordered state in U$_2$Zn$_{17}$ is not easily suppressed, but is stabilized under high pressures up to 8.7 GPa. Also, the antiferromagnetic state in UCu$_5$ is stabilized under high pressure. These results suggest that the pressure effect on the antiferromagnetic state in uranium heavy fermion compounds is highly different from those of heavy fermion cerium compounds.

Acknowledgements

This work was financially supported by the Grant-in-Aid for Creative Scientific Research (15GS0213), Scientific Research of Priority Area and Scientific Research (A, B and C) from the Ministry of Education, Culture, Sports, Science and Technology (MEXT) and Japan Society of the Promotion of Science (JSPS).

References

[1] Flouquet J 2006 Prog. Low Temp. Phys. 15 139
[2] Löhneysen H v, Rosch A, Vojta M, Wölle P 2007 Rev. Mod. Phys. 79 1015
[3] Ott H R, Rudigier H, Delsing P and Fisk Z 1984 Phys. Rev. Lett. 52 1551
[4] Fischer H E, Swartz E T, Pohl R O, Jones B A, Wilkins J W, and Fisk Z 1987 Phys. Rev. B 36 5330
[5] Cox D E, Shirane G, Shapiro S M, Appli G, Fisk Z, Smith J L, Kjems J and Ott H R 1986 Phys. Rev. B 33 3614
[6] Dunstan D J and Spain I L 1989 J. Phys. E: Sci. Instrum. 22 913
[7] Spain I L and Dunstan D J 1989 J. Phys. E: Sci. Instrum. 22 923
[8] Bell P M and Mao H K 1981 Carnegie Inst. Wash. Yearb. 80 404
[9] Zha C S, Mao H K and Hemley R J 2000 Proc. Natl Acad. Sci. USA 97 13494
[10] Thompson J D, Fisk Z and Ott H R 1986 J. Magn. Magn. Mat. 54-57 393
[11] Thompson J D, Lawrence J M 1994 Handbook on the Physics and Chemistry of Rare Earths, Vol. 19, chap.133, ed. Gschneider K A Jr and Eyring K (North-Holland)
[12] Ott H R, Rudigier H, Felder E, Fisk Z and B. Batlogg 1985 Phys. Rev. Lett. 55 1595
[13] Nakashima M, Sugitani I, Okuda Y, Shishido H, Matsuda T D, Haga Y, Hedo M, Uwatoko Y, Settai R and Onuki Y 2006 Frontiers of basic science towards new physics earth and space science mathematics, p. 267, eds. Takabe H, Luong N H and Onuki Y (Osaka University Press)
[14] Palstra T T M, Menovsky A A and Mydosh J A 1986 Phys. Rev. B 33 6527
[15] Raymond S and Jaccard D 2000 Phys. Rev. B 61 8679
[16] Kodowaki K and Woods S B 1986 Solid State Commun. 58 507
[17] Migliori A, Sarrao J L, Mandrus D, Fisk Z, Balatsky A, Trugman S A, Thompson J D, Maple M B 1994 Physica B 199k 200 36
[18] Kagayama T, Oomi G, Iki K, Mori N, Onuki Y and Komatsubara T 1994 Journal of Alloys and Compounds 213-214 387
[19] Kagayama T, Oomi G, Ito E, Onuki Y and Komatsubara T 1994 J. Phys. Soc. Jpn. 63 3927
[20] Vedel I, Redon A M, Mignot J -M, Leger J M 1987 J. Phys. F: Met. Phys. 17 849
[21] Kumar R S, Cornelius A L and Sarrao J L 2004 Phys. Rev. B 70 214526
[22] 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 394 39
[23] Hegger H, Petrovic C, Moshopoulou E G, Hundley M F, Sarrao J L, Fisk Z and Thompson: J D 2000 Phys. Rev. Lett. 84 4986