Pressure-induced novel superconductivity and heavy fermion state in rare earth compounds

Fuminori Honda¹, Yusuke Hirose¹, Shingo Yoshiuchi¹, Shinichi Yasui¹, Tetsuya Takeuchi², Ismardo Bonalde³,⁴, Katsuya Shimizu³, Rikio Settai¹, and Yoshichika Ônuki¹

¹Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan
²Low Temperature Center, Osaka University, Toyonaka, Osaka 560-0043, Japan
³Centro de Física, Instituto Venezolano de Investigaciones Científicas, Apartado 20632, Caracas 1020-A, Venezuela
⁴KYOKUGEN, Center for Quantum Science and Technology under Extreme Conditions, Osaka University, Osaka, 560-8531, Japan
E-mail: honda@phys.sci.osaka-u.ac.jp

Abstract. We have carried out the electrical resistivity measurements under high pressures up to 24 GPa for CeTX₃ (T: Co and Ir, X: Si and Ge), CePd₅Al₂ and YbIr₂Zn₂₀ in order to investigate quantum criticality and superconductivity. Antiferromagnets CeTX₃ with the non-centrosymmetric tetragonal structure show superconductivity under high pressures and reveal a huge upper critical field for \( H \parallel [001] \). An antiferromagnet CePd₅Al₂, which is an isostructural family of a heavy fermion superconductor NpPd₅Al₂, also shows superconductivity under high pressures. In these compounds, superconductivity appears in the vicinity of quantum critical point. On the other hand, YbIr₂Zn₂₀ without magnetic ordering approaches to the quantum critical point with increasing pressure, and exhibits a super-heavy fermion state exceeding 10 J/(K²·mol).

1. Introduction
The interplay of magnetism and superconductivity has attracted a great interest in \( f \)-electron systems of Ce and U compounds [1, 2]. Recently, it is widely recognized that pressure is a useful method to change the electronic state in these compounds. When pressure \( P \) is applied to an antiferromagnet, the Néel temperature \( T_N \) decreases with increasing pressure and becomes zero at a critical pressure \( P_c \): \( T_N \to 0 \) for \( P \to P_c \). An electronic state at \( P_c \) is called the quantum critical point. Heavy fermion superconductivity and the non-Fermi liquid nature are often observed in this critical pressure region or in the vicinity of the quantum critical point.

In this paper, we report characteristic heavy fermion superconducting properties in CeTX₃ (T: Co and Ir, X: Si and Ge) and CePd₅Al₂. Both compounds are antiferromagnets at ambient pressure, but become superconductive under pressure. A huge upper critical field \( H_{c2} \) for \( H \parallel [001] \) is characteristic in CeTX₃. Pressure also induces a super-heavy fermion state for YbIr₂Zn₂₀ without magnetic ordering.

2. Experimental Results and Discussion
2.1. CeTX₃
CeTX₃ (T: transition metal, X: Si, Ge) compounds crystallize in the BaNiSn₃-type tetragonal structure without inversion symmetry along the [001] direction, as shown in inset of Fig. 1. Most of CeTX₃
order antiferromagnetically at ambient pressure. Figure 1 shows the Néel temperature with respect to the averaged lattice parameter. This corresponds to the Doniach phase diagram [3], which is based on the competition between the RKKY interaction and the Kondo effect. The RKKY interaction enhances the magnetic ordering, while the Kondo effect quenches the magnetic moment of the 4f electrons.

\[ T_N = \frac{1}{\sqrt{a^2 c}} \, \text{(Å)} \]

\[ \sqrt{a^2 c} \]

Figure 1. Néel temperature \( T_N \) with respect to the averaged lattice parameter, \( \sqrt{a^2 c} \), in noncentrosymmetric compounds CeTX3. Inset shows the tetragonal crystal structure of CeTX3.

First we studied an effect of pressure to an electronic state of CeIrGe3 with a Néel temperature \( T_N = 8.7 \) K. Figure 2(a) shows the temperature dependence of the electrical resistivity under high pressures up to 24 GPa. The current \( J \) is applied along the [100] direction. With increasing pressure, the electrical resistivity indicates a typical heavy fermion behavior at 17 and 20 GPa as in CeCu6 [4], revealing a resistivity peak at 50 and 100 K, respectively.

At 20 GPa, the electrical resistivity indicates a sudden drop below \( T_{sc} = 1.5 \) K, which is attributed to the superconducting transition. The superconducting state coexists with the antiferromagnetic state at 20 GPa, and only a superconducting transition occurs at 24 GPa. The resistivity at 24 GPa follows a \( T \)-linear dependence in the temperature range from 1.7 to 70 K, indicating that the system becomes a non-Fermi liquid (NFL) state. This behavior is also found in CeCoGe3 and CeIrSi3.

Figure 2(b) shows a pressure-temperature phase diagram of CeIrGe3. The Néel temperature \( T_N \) decreases with two steps: the first step is located around 10 GPa and the second step is about 21 GPa, and superconductivity appears above 20 GPa, with \( T_{sc} = 1.5 \) K. The step-like change in \( T_N \) implies a change of magnetic structures as in CeCoGe3 [5]. An ordered moment in CeCoGe3 is oriented along the [001] direction, with \( q = (0 \ 0 \ 1/2) \) in CeCoGe3 [6], and this magnetic structure is most likely not favorable for superconductivity, especially in the tetragonal structure. The corresponding magnetic structure of CeCoGe3, together with CeIrGe3, might be changed as a function of pressure, and superconductivity might be realized in favorable magnetism.
We show in Figs. 3(a) and 3(b) the similar pressure-temperature phase diagram in CeCoGe$_3$ and CeIrSi$_3$, respectively. As expected from the result in Fig. 1, the critical pressure $P_c$, where the Néel temperature merges in the superconducting transition temperature $T_{sc}$, is $P_c = 5.8$ GPa in CeCoGe$_3$ [5, 7] and 2.25 GPa in CeIrSi$_3$ [8], which are compared with $P_c = 22$ GPa in CeIrGe$_3$. It is characteristic that a pressure $P_{c*}$, where the $T_{sc}$ value becomes maximum, is not $P_c$, but is located in the non-magnetic electronic state, separated from $P_c$. $P_{c*}$ is 6.5 GPa in CeCoGe$_3$ and 2.6 GPa in CeIrSi$_3$.

A huge upper critical field $H_{c2}$ value for $H \parallel [001]$ is characteristic in CeTX$_3$, as shown in Fig. 4. The quantum critical behavior, non-centrosymmetry, and a strong-coupling nature of superconductivity are combined into the huge $H_{c2}(0)$ value for $H \parallel [001]$: $H_{c2} \approx 45$ T for $H \parallel [001]$, while $H_{c2} \approx 9.5$ T for $H \parallel [110]$ in CeIrSi$_3$ [8]. It is noted that the Fermi surface is expected to be split into two Fermi surfaces based on the antisymmetric spin-orbit interaction, and the corresponding spins of conduction electrons are rotated clockwise or anti-clockwise, depending on the up and down spin states. Therefore, a Pauli paramagnetic effect in superconductivity does not occur for $H \parallel [001]$ because the magnetic field is perpendicular to the directions of all the spins. It is stressed that the $H_{c2}$ value becomes maximum at $P_{c*}$ in CeTX$_3$.

2.2. CePd$_5$Al$_2$

Superconductivity has been observed in rare earth, uranium and nowadays even transuranium compounds. It was, however, a surprise that heavy fermion superconductivity was discovered in NpPd$_5$Al$_2$ [10], since most of Np compounds order magnetically. CePd$_5$Al$_2$, which is an analog of a heavy fermion superconductor NpPd$_5$Al$_2$, is an antiferromagnet with the Néel temperature $T_N = 4$ K. It is noted that these compounds crystallize in the layered tetragonal structure, revealing a quasi-two dimensional electronic state [10, 11, 12].
The antiferromagnetic state of CePd$_5$Al$_2$ is also changed into the superconducting state at about 10 GPa. We show in Fig. 5(a) the upper critical field $H_{c2}$, together with that for NpPd$_5$Al$_2$ at ambient pressure, as shown in Fig. 5(b).

The slope of $H_{c2}$ at $T_{sc} = 0.9$ K and the $H_{c2}$ value at 0 K are obtained as $-dH_{c2}/dT = 1.8$ T/K at and $H_{c2}(0) = 0.4$ T for $H \parallel [001]$, and $-dH_{c2}/dT = 10$ T/K and $H_{c2}(0) = 0.68$ T for $H \parallel [100]$ in CePd$_5$Al$_2$. It is noted that the orbital limiting field $H_{c2}(0) = 0.73 \left(-dH_{c2}/dT\right)T_{sc}$ for the clean limit is obtained to be 6.6 T for $H \parallel [100]$, which is much larger than the experimental value of $H_{c2}(0) = 0.68$ T. A large $H_{c2}$ value is expected for $H \parallel [100]$, compared with the $H_{c2}$ value for $H \parallel [001]$, because the Fermi surface is nearly cylindrical and the the cyclotron effective mass for $H \parallel [100]$ is much larger than that for $H \parallel [001]$ [13]. It is clear that the upper critical field is suppressed by the strong paramagnetic effect.

On the other hand, the $H_{c2}$ value for $H \parallel [001]$ is larger than that for $H \parallel [100]$ in NpPd$_5$Al$_2$, as shown in Fig. 5(b). The reason is not clear, but the paramagnetic effect might be anisotropic because the anisotropy of magnetic susceptibility for $H \parallel [100]$ and [001] is different between two compounds.

2.3. YbIr$_2$Zn$_{20}$

YbIr$_2$Zn$_{20}$ (T: transition metal) compounds crystallize in the caged cubic CeCr$_2$Al$_{20}$ (Fd$ar{3}$m)-type structure with a lattice parameter of about 14 Å [14], where Yb and T atoms locate in the fcc diamond lattice and pyrochrore structure, respectively, and are surrounded by Zn atoms. Since the Yb-Yb distance is quite large, 6 Å, the magnetic exchange interaction between Yb atoms becomes weak. In fact, YbIr$_2$Zn$_{20}$ compounds do not order magnetically. The corresponding electronic specific heat coefficient $\gamma$ is very large: 540 mJ/(K$^2$-mol) in YbIr$_2$Zn$_{20}$ and 8000 mJ/(K$^2$-mol) in YbCo$_2$Zn$_{20}$ [14].

An effect of pressure on the electronic state of Yb compounds is, however, highly different from that of Ce compounds. Since the volume of the lattice becomes smaller when the valence changes from the non-magnetic Yb$^{2+}$ towards magnetic Yb$^{3+}$. Applying pressure drives the system from a non-magnetic state to a magnetic state. For example, a non-magnetic Yb compound or a so-called valence fluctuating compound YbCu$_2$Si$_2$ is known to order magnetically at pressures higher than 8 GPa [15].

Recently, we have found a non-linear increase of magnetization or a metamagnetic behavior at $H_m \approx 10$ T in YbIr$_2$Zn$_{20}$ at temperatures smaller than $T_{x_{\text{max}}} = 7.4$ K where the magnetic susceptibility indicates a maximum [16]. Based on the Doniach phase diagram, it is expected that YbIr$_2$Zn$_{20}$ reaches the quantum critical point and finally indicates magnetic ordering at high pressures. Note that there is discovered no pressure-induced superconductor in Yb compounds.

Figure 6(a) shows the transverse magnetoresistance in YbIr$_2$Zn$_{20}$ under pressures up to 5.5 GPa. A shoulder-like anomaly at $H_m = 9.7$ T under 0 GPa corresponds to the metamagnetic behavior [17]. With increasing pressure, $H_m$ shifts to lower magnetic fields, and the resistivity anomaly at $H_m$ becomes sharp and distinct. The metamagnetic behavior is found at $H_m = 0.15$ T at 5.0 GPa but no trace of the
Figure 6. (a) Magnetic field dependence of (a) the electrical resistivity at 0.1 K and (b) $A$-value under high pressure in YbIr$_2$Zn$_{20}$.

metamagnetic anomaly has been observed at 5.5 GPa. The critical pressure is thus suited between 5.0 and 5.5 GPa.

The low-temperature resistivity shows a quadratic temperature dependence of $\rho = \rho_0 + AT^2$ ($\rho_0$: residual resistivity) at 0 T and in magnetic fields, reflecting the Fermi liquid nature. In Fig. 6(b), we show the magnetic field dependence of the $A$ coefficients at different pressures. It is clear that a broad peak at $H_m = 9.7$ T under 0 GPa is changed into a distinct peak with increasing pressure, together with an anomalous enhancement of the $A$ value at high pressures. The $A$ value reaches about 380 $\mu$Ω·cm/K$^2$ at 5.0 GPa, which corresponds to 15 J/(K$^2$·mol) based on the so-called grand-Kadowaki-Woods relation with degeneracy of quasiparticles $N = 4$ [18]. This is the highest value ever obtained in the heavy fermion systems. At 5.5 GPa, the $A$ value decreases monotonically with increasing the magnetic field. The critical pressure is thus obtained as $P_c \approx 5.2$ GPa. At pressures larger than $P_c \approx 5.2$ GPa, an antiferromagnetic ordered state was observed at 7.6 and 8.6 GPa. We show in Fig. 7 the pressure dependence of the metamagnetic field $H_m$ and the Néel temperature $T_N$. Note that no trace of superconductivity has been observed at 5.0 and 5.5 GPa down to 80 mK.

Figure 7. Pressure dependence of the metamagnetic field $H_m$ (•) and the Néel temperature $T_N$ (■) in YbIr$_2$Zn$_{20}$.

3. Summary
We measured the electrical resistivity in antiferromagnets CeTX$_3$ and CePd$_5$Al$_2$ with the tetragonal structure, together with a cubic non-magnetic Yb-compound YbIr$_2$Zn$_{20}$ under high pressures. The experimental results are summarized as follows:
1) Pressure-induced superconductivity was found under high pressures in CeIrGe, CeCoGe, and CeIrS3. The characteristic pressure P^c in with the highest T_c and H_c2 values are about 24, 6.5, and 2.6 GPa, respectively. A huge H_c2 value for H|| [001] is characteristic in these compounds with the non-centrosymmetric crystal structure.

2) An antiferromagnet CePd3Al2 becomes superconductive above 9.0 GPa. Anisotropy of H_c2 between H|| [100] and [001] is reasonable from the quasi-two dimensional Fermi surface, which is, however, highly different from anisotropy of H_c2 in NpPd3Al2.

3) The critical pressure in YbIr2Zn20 is determined to be 5.2 GPa. A huge A value of a Fermi liquid relation ρ = ρ0 + AT^2 is obtained as A = 380 μΩ-cm/K^2 at 5.0 GPa, which corresponds to 15 J/(K^2·mol). The super-heavy fermion state is realized in YbIr2Zn20 under pressures.

Acknowledgments
This work was supported by a Grant-in-Aid for Scientific Research on Specially Promoted Research (No.20001004), Osaka University: Global COE Program “Core Research and Engineering of Advanced Materials-Interdisciplinary Education Center for Materials Science” (No.G10), Grants-in-Aid for Scientific Research on Innovative Areas “Heavy Electrons” (20102002, 23102715), Priority Areas of New Materials Science Using Regulated Nano Spaces (22013009), and Scientific Research (B)(21340102) from the Ministry of Education, Culture, Sports, Science and Technology, Japan. This work was also supported by the “Exciting Leading-Edge Research Project” at Osaka University.

References
[1] Ōnuki Y, Settai R, Sugiyama K, Takeuchi T, Kobayashi T C, Haga Y, and Yamamoto E 2004 J. Phys. Soc. Jpn. 73 769
[2] Pfleiderer C 2009 Rev. Mod. Phys. 81 1551
[3] Doniach S, Valence Instabilities and Related Narrow Band Phenomena, ed. Parks R D (Plenum, New York, 1977), p. 169
[4] Ōnuki Y and Komatsubara T 1987 J. Magn. Magn. Mater. 63 & 64 281.
[5] Knebel G, Aoki D, Lapertot G, Salce B, Flouquet J, Kawai T, Muranaka H, Settai R, and Ōnuki Y 2009 J. Phys. Soc. Jpn. 78 074714
[6] Kaneko K, Metoki N, Takeuchi T, Matsuda T D, Haga Y, Tamizhavel A, Settai R, Ōnuki Y 2009 J. Phys. Conf. ser. 150 042082
[7] Kawai T, Muranaka H, Measson M A, Shimoda T, Doi Y, Matsuda T D, Haga Y, Knebel G, Lapertot G, Aoki D, Flouquet J, Takeuchi T, Settai R, and Ōnuki Y 2007 J. Phys. Soc. Jpn. 76 064716
[8] Settai R, Miyauchi Y, Takeuchi T, Levy F, Sheikin I, and Ōnuki Y 2008 J. Phys. Soc. Jpn. 77 073705
[9] Tada Y, Kawakami N, and Fujimoto S 2008 Phys. Rev. Lett. 101 267006
[10] Aoki D, Haga Y, Matsuda T D, Tateiwa N, Ikeda S, Homma Y, Sakai H, Shiokawa Y, Yamamoto E, Nakamura A, Settai R, and Ōnuki Y 2007 J. Phys. Soc. Jpn. 76 063701
[11] Yamagami H, Aoki D, Haga Y, and Ōnuki Y 2007 J. Phys. Soc. Jpn. 76 083708
[12] F. Honda, Measson M A, Nakano Y, Yoshitani N, Yamamoto E, Haga Y, Takeuchi T, Yamagami H, Shimizu K, Settai R, and Ōnuki Y 2008 J. Phys. Soc. Jpn. 77 043701
[13] Nakano Y, Honda, Takeuchi T, Sugiyama K, Hagiwara M, Kindo K, Yamamoto E, Haga Y, Settai R, Yamagami H, and Ōnuki Y 2010 J. Phys. Soc. Jpn. 79 024702
[14] Torkachvili M S, Jia S, Mun E D, Hannans S T, Black R C, Neils W K, Martien D, Bud’ko S L, and Canfield P C 2007 Proc. Natl. Acad. Sci. U.S.A. 104 9960
[15] Winkelmann H, Abd-Elmeguid M M, Micklitz H, Sanchez J P, Vulliet P, Alami-Yadri K, and Jaccard D 1999 Phys. Rev. B 60 3324
[16] S. Yoshitani, M. Toda, M. Matsushita, S. Yasui, Y. Hirose, M. Ohya, K. Katayama, F. Honda, Sugiyama K, Hagiwara M, Kindo K, Takeuchi T, Yamamoto E, Haga Y, Settai R, T. Tanaka, Kubo Y, Onuki Y 2009 J. Phys. Soc. Jpn. 78 123711
[17] Takeuchi T, S. Yasui, M. Toda, M. Matsushita, S. Yoshitani, M. Ohya, K. Katayama, Y. Hirose, Yoshitani N, F. Honda, Sugiyama K, Hagiwara M, Kindo K, Yamamoto E, Haga Y, T. Tanaka, Kubo Y, Settai R, and Ōnuki Y 2010 J. Phys. Soc. Jpn. 79 064609
[18] Tsuji N, Kontani H, Yoshimura K 2005 Phys. Rev. Lett. 94 057201