Characteristic magnetic phase diagram in heavy-fermion compound $\text{YbCo}_2\text{Zn}_{20}$

T Takeuchi$^{1,3}$, S Yoshiuchi$^2$, M Ohya$^2$, M Matsushita$^2$, S Yasui$^2$, Y Hirose$^2$, K Sugiyama$^{2,3}$, F Honda$^2$, R Settai$^2$ and Y Ônuki$^2$

1. Introduction

Yb-based compounds have been extensively investigated since the Yb$^{3+}$ has one 4$f$-hole and are considered as a countersystem to one-4$f$-electron Ce-based compounds. The electron-hole analogy between Yb$^{3+}(4f^{13})$ and Ce$^{3+}(4f^1)$ electronic configurations tends to induce the localization of 4$f$ electrons, enhancing a magnetically ordered state when pressure is applied to nonmagnetic Yb-based compounds. Namely, the pressure reduces the unit cell volume, which leads to the trivalent magnetic Yb$^{3+}(4f^{13})$ state rather than to the divalent nonmagnetic Yb$^{2+}(4f^{14})$ state. Another interesting feature of Yb-based compounds is valence transition or fluctuation due to the small energy gap for Yb$^{3+}(4f^{13}) - Yb^{2+}(4f^{14})$ charge instabilities. While the number of cerium compounds showing such anomalous behavior is fairly large in number, there are relatively few Yb-based compounds showing typical properties of valence fluctuation or heavy-fermion behavior. This may be due to a relatively smaller spatial extension of the 4$f$ shell in Yb$^{3+}$ ion as compared to that in the Ce$^{3+}$ ion. Namely, the smaller extension of the 4$f$ shell in Yb-based compounds leads to the localization of 4$f$ electrons and hence smaller hybridization between 4$f$ electrons and conduction electrons. It is therefore needed to identify new Yb-based compounds and study their low-temperature properties.

Recently, a new family of Yb-based heavy-fermion compounds YbT$_2$Zn$_{20}$ (T = Fe, Co, Ru, Rh, Os, and Ir) with the cubic CeCr$_2$Al$_{20}$-type ($Fd\bar{3}m$ space group) structure has been reported [1]. The lattice constant of these compounds is very large, i.e., $a \sim 14$ Å, and hence the distance between Yb-Yb atoms is large, more than 6 Å. In YbT$_2$Zn$_{20}$, the Yb atom forms the diamond
structure and is encapsulated in the Frank-Kasper cage formed by 16 zinc atoms, while the transition atom T has the icosahedral zinc coordination, forming caged structures. These compounds show no long-range magnetic ordering at temperatures down to 0.02 K, and the electronic specific heat coefficient $\gamma$ at low temperatures was found to be very large, ranging between 500 and 800 mJ/(K$^2$-mol) [1]. For YbCo$_2$Zn$_{20}$, YbRb$_2$Zn$_{20}$, and YbIr$_2$Zn$_{20}$, we succeeded in growing single crystals and found an abrupt nonlinear increase of magnetization or the metamagnetic behavior in the nonmagnetic heavy-fermion state [2, 3, 4]. The metamagnetic anomaly was observed at $H_m = 6$, 63, and 97 kOe below $T_{\chi_{\text{max}}}$ = 0.32, 5.8, and 7.4 K for YbCo$_2$Zn$_{20}$, YbRb$_2$Zn$_{20}$, and YbIr$_2$Zn$_{20}$, respectively. Here, $T_{\chi_{\text{max}}}$ is the temperature at which the temperature dependence of magnetic susceptibility $\chi$ shows a local broad maximum.

Since the magnetic susceptibility of YbCo$_2$Zn$_{20}$ increases continuously with decreasing temperature in the temperature range from room temperature down to 2 K, following the Curie-Weiss law, the Kondo temperature in YbCo$_2$Zn$_{20}$ is expected to be much lower than that in YbIr$_2$Zn$_{20}$ with $T_{\chi_{\text{max}}}$ = 7.4 K ($T_K$ = 21 K) [1]. The corresponding electronic specific heat coefficient of YbCo$_2$Zn$_{20}$ is extremely large: $\gamma \approx 8 J/(K^2\text{-mol})$ at low temperatures [1]. The cyclotron effective mass at zero magnetic field was estimated to be 100-500$m_0$ from the magnetic field dependences of the coefficient $\sqrt{A}$ in the Fermi liquid relation of the electrical resistivity, $\rho = \rho_0 + AT^2$, and cyclotron effective mass determined from the dHvA experiment [4]. The extremely large $A = 160 \mu\Omega\cdot\text{cm}/K^2$ at $H = 0$ kOe was found to be reduced largely with increasing magnetic field to $A = 0.021 \mu\Omega\cdot\text{cm}/K^2$ at 150 kOe. Here, we note that the coefficient $\sqrt{A}$ correlates with the $\gamma$ value in the Fermi liquid relation.

In this paper, we present the characteristic $H$-$T$ phase diagrams of the heavy-fermion compound YbCo$_2$Zn$_{20}$ obtained from specific heat, ac-susceptibility, and electrical resistivity measurements under various magnetic fields.

2. Experimental Procedure

Single crystal samples of YbCo$_2$Zn$_{20}$ were grown by the same Zn-self flux method as that used for YbIr$_2$Zn$_{20}$ [2, 3]. The specific heat was measured by the quasi-adiabatic heat pulse method under magnetic fields up to 90 kOe. The electrical resistivity and magnetoresistance were measured by an ordinary four-probe dc and/or ac method under magnetic fields of up to 150 kOe. The ac-susceptibility was measured by the mutual inductance method. A dilution refrigerator was utilized for the measurement below 1 K.

3. Experimental results and discussion

In a low magnetic field region, the magnetic field dependence of $\chi_{\text{ac}}$ shows a broad peak at $H_m = 6$ kOe at temperatures below $T_{\chi_{\text{max}}} = 0.32$ K, as shown in figure 1(b), which corresponds to the metamagnetic behavior of magnetization. As shown in figure 1(c), the temperature dependence of $\chi_{\text{ac}}$ at $H = 0$ kOe shows a broad peak at $T_{\chi_{\text{max}}} = 0.32$ K and decreases slightly at lower temperatures. $T_{\chi_{\text{max}}}$ shifts to lower temperatures when the magnetic field is increased and disappears around $H_m = 6$ kOe. For further increasing the magnetic field, a broad peak in $\chi_{\text{ac}}$ appears again above $H_m$ and shifts to higher temperatures, as indicated by open circles in figure 1(a). Interestingly, the magnetic field dependence of the specific heat in the form of $C/T$ at 0.095 K shows distinct two peaks at about 4.0 and 7.5 kOe, as indicated by arrows in figure 1(d), and exhibits a local minimum at $H_m$. With increasing temperature, the peak at a higher (lower) magnetic field shifts to higher (lower) fields, as indicated by arrows for the data at 0.2 K. These points are also plotted in figure 1(a) by open triangles.

Evolution of the temperature dependence of electrical resistivity under magnetic fields is also shown in figure 1(a) as the contour plot of the power law exponent $n$ for the temperature dependent resistivity in the form of $\rho^n$. This plot was constructed by interpolating the results of temperature sweeps at sixteen values of applied magnetic fields: a 1 kOe interval from 0
to 15 kOe. From this plot, it is found that there are two crossover lines below and above the metamagnetic anomaly at \( H_m = 6 \) kOe. The Fermi-liquid state develops at lower temperatures than these crossover lines. Note that these characteristic features at low magnetic fields, including the metamagnetic behavior at \( H_m \), are almost the same for the three principal field directions, \( H \parallel \langle 100 \rangle \), \( \langle 110 \rangle \), and \( \langle 111 \rangle \).

In contrast to the low field behavior, the field-induced ordered phase (FIOP) was observed in a limited angular range around the \( \langle 111 \rangle \) direction above the transition field \( H_Q = 60 \) kOe below \( T_Q = 0.6 \) K, as shown in figure 2(a). At the phase boundary of the FIOP, the electrical resistivity and specific heat show a clear anomaly at \( H_Q \) or \( T_Q \), as shown in figures 2(b) and 2(c). At \( T = 0.08 \) K, the magnetoresistance shows peaks at \( H_m = 6 \) kOe and \( H_Q = 60 \) kOe, which correspond to the metamagnetic anomalies. With increasing temperature, the peak at \( H_m \) rapidly smears out and becomes obscure at a temperature between 0.30 and 0.45 K, which is similar to the results for \( H \parallel \langle 100 \rangle \) [4]. The peak at \( H_Q \), on the other hand, slightly shifts to higher fields and becomes broader with increasing temperature.

The specific heat at 50 kOe, which is below \( H_Q = 60 \) kOe, decreases linearly below 1 K without any anomalous behavior. When we apply the magnetic field of 70 kOe, a \( \lambda \)-shaped peak was observed at 0.4 K, indicating the evolution of FIOP. The \( \lambda \)-shaped peak shifts to about 0.6 K and the peak becomes larger at the magnetic field of 90 kOe. These results strongly support the existence of the field-induced long-range-ordered phase for \( H \parallel \langle 111 \rangle \). Very recently, the FIOP of YbCo\(_2\)Zn\(_2\)O\(_4\) for \( H \parallel \langle 111 \rangle \) has been confirmed by the capacitance magnetization measurement at low temperatures [5].

The characteristic features of the FIOP in YbCo\(_2\)Zn\(_2\)O\(_4\) are very similar to those in PrFe\(_4\)P\(_{12}\), where the FIOP was observed in a limited angular range around the \( \langle 111 \rangle \) direction above the other ordered phase [6]. The FIOP in PrFe\(_4\)P\(_{12}\) has been explained by considering a two-
Figure 2. (a) $H$-$T$ phase diagram for $H \parallel \langle 111 \rangle$ in YbCo$_2$Zn$_2$0. Squares are determined by the specific heat measurement, and triangles are obtained by the resistivity measurement. (b) Magnetoresistances with the current $J \parallel \langle 110 \rangle$ for $H \parallel \langle 111 \rangle$ at several selected temperatures, and (c) temperature dependences of the specific heat $C$ under several magnetic fields along the $\langle 111 \rangle$ direction.

sublattice mean-field model with quadrupole interactions for $O^0_2$ and $O^2_2$. In this case, the level crossing of the two lowest levels occurs for $H \parallel \langle 111 \rangle$ in the proposed CEF scheme. The numerical analyses for YbCo$_2$Zn$_2$0 based on the CEF model suggest that the FIOP for $H \parallel \langle 111 \rangle$ is the antiferro-quadrupolar ordered phase with the quadrupole moment of $O^0_2$ or $O^2_2$. In order to clarify the order parameter of the FIOP, a further experimental study is necessary.

4. Conclusion

$H$-$T$ phase diagrams of the heavy-fermion compound YbCo$_2$Zn$_2$0 were studied precisely using single crystal samples. The characteristic crossover phase diagram was obtained for the low field region below about 15 kOe, which was found to be independent of the direction of magnetic field. On the other hand, the FIOP was observed in a limited angular range around the $\langle 111 \rangle$ direction above the transition field $H_Q = 60$ kOe in the temperature range below $T_Q = 0.6$ K.

Acknowledgments

This work was supported by a Grant-in-Aid for Scientific Research on Innovative Areas (No. 20102002), Specially Promoted Research (No. 20001004), Young Scientists (B), (No. 18740219) and Osaka University Global COE Program (G10) from the Ministry of Education, Culture, Sports, Science, and Technology, Japan.

References

[1] Torikachvili 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. U.S.A. 104 9960
[2] Yoshiuchi S et al 2009 J. Phys. Soc. Jpn. 78 123711
[3] Takeuchi T et al 2010 J. Phys. Soc. Jpn. 79 064609
[4] OhyA M, Matsushita M, Yoshiuchi S, Takeuchi T, Honda F, Settai R, Tanaka T, Kubo Y and Ōnuki Y 2010 J. Phys. Soc. Jpn. 79 083601
[5] Shimura Y, Sakakibara T, Yoshiuchi S, Honda F, Settai R and Ōnuki Y 2011 J. Phys. Soc. Jpn. 80 073707
[6] Tayama T, Custers J, Sato H, Sakakibara T, Sugawara H and Sato H 2004 J. Phys. Soc. Jpn. 73 3258