Pressure-induced Magnetic Transition in a Single Crystal of $YbCo_2Zn_{20}$

Yuta SAIGA, Kazuyuki MATSUBAYASHI[1], Tetsuya FUJIWARA[2], Masashi KOSAKA, Susumu KATANO, Masato HEDO[3], Takehiko MATSUMOTO[1], and Yoshiya UWATOKO[1]

Graduate School of Science and Engineering, Saitama University, Saitama 338-8570, Japan
[1]*Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan*
[2]*Graduate School of Science and Engineering, Yamaguchi University, Yamaguchi 753-8512, Japan*
[3]*Faculty of Science, University of the Ryukyus, Nishihara, Okinawa 903-0213, Japan*

We have measured the electrical resistivity of a single crystal of $YbCo_2Zn_{20}$ at pressures up to 2.37 GPa and at temperatures from 50 mK to 300 K. Above a critical pressure P_c ($\sim$ 1 GPa), we have found a resistivity anomaly at T_M ($\sim$ 0.15 K at 1 GPa) that increases with the pressure. At the ambient pressure, the system shows a nonmagnetic ground state described by the Fermi-liquid model. The T^2 coefficient of the electrical resistivity A strongly increases with the pressure upon approaching P_c. However, in the vicinity of P_c, the temperature dependence of the resistivity deviates from the Fermi-liquid description. These observations suggest that the application of hydrostatic pressure induces a magnetically ordered state for $P \geq P_c$ and $T \leq T_M$.

KEYWORDS: $YbCo_2Zn_{20}$, heavy fermion, pressure measurement, pressure-induced magnetic transition, QCP

In intermetallic compounds, including Ce and Yb, the hybridization between the $4f$ and itinerant conduction-band electrons induces the instability of magnetic moments and charge configurations. In recent years, one of the most interesting topics is the ground state properties of heavy fermion metals located at or close to a magnetic quantum critical point (QCP).[1)] The application of external pressure is one of the important tools for controlling the electronic configurations as well as chemical pressure. In the case of heavy fermion Ce compounds exhibiting antiferromagnetic order, such as $CeIn_3$ and $CePd_2Si_2$,[2)] the magnetic order is suppressed by applying pressure. Interestingly, unconventional superconductivity appears in the vicinity of the QCP at which the magnetic ordering temperature is decreased to zero.

So far, pressure-induced magnetic transitions have been observed in some Yb-based compounds.[3–7)] The key point here is that Yb ions fluctuate between the nonmagnetic Yb^{2+} ($J = 0$) and the magnetic Yb^{3+} ($J = 7/2$) states. Since the ionic volume of the magnetic Yb^{3+} state is smaller than that of the nonmagnetic Yb^{2+} one, applying pressure stabilizes the magnetic Yb^{3+} configuration and induces the appearance of a magnetically ordered state in contrast to the Ce case. However, the critical pressure for these compounds is as high as or higher than 6 GPa, which prevents us from understanding the physics in the vicinity of the magnetic QCP because of difficulties in high-pressure experiments.

The series of compounds YbT_2Zn_{20} (T = Fe, Co, Ru, Rh, Or, Ir) belongs to a new heavy fermion system crystallizing in the cubic $CeCr_2Al_{20}$ structure.[8,9)] All these compounds show an enhanced Sommerfeld coefficient of the specific heat γ exceeding 400 mJ/mol K^2. The high-temperature magnetic susceptibility of these compounds follows the Curie-Weiss law with the effective moments close to the value for the free Yb^{3+} ion ($\mu_{eff} = 4.54\ \mu_B$), although there is no indication of magnetic order down to 20 mK. In the YbT_2Zn_{20} family, $YbCo_2Zn_{20}$ exhibits some notable features as follows. The low temperature electrical resistivity and specific heat can be described by the formulas $\rho = \rho_0 + AT^2$ and $C/T = constant$, as expected from the Fermi-liquid behavior; the values of A and γ are estimated to be $\sim$165 $\mu\Omega$ cm/K^2 and $\sim$7900 mJ/mol K^2, respectively.[8)] The large values of A and γ are due to the small Kondo temperature T_K, which is estimated to be several Kelvin. If the ground state properties in this compound are dominated by the competition between the RKKY interaction and the Kondo interaction, the system is located near a magnetic instability. Hence, it appears that $YbCo_2Zn_{20}$ is an interesting system due to its proximity to the QCP and it is expected to cross the magnetic QCP by applying pressure.

In this letter, we present the first report on the effect of pressure on $YbCo_2Zn_{20}$. We prepared single crystals of $YbCo_2Zn_{20}$ and measured the electrical resistivity under pressures up to 2.37 GPa and at temperatures from 50 mK to 300 K.

Single crystal samples of $YbCo_2Zn_{20}$ were prepared by the Zn self-flux method. The starting elements were mixed in the ratio of Yb:Co:Zn = 1:2:30 and placed in an Al_2O_3 crucible in an evacuated quartz ampoule. The sample was heated up to 900 °C, maintained at that temperature for 2 h, and then slowly cooled to 750 °C at the rate of 1 °C/h. At 750 °C, the excess flux was removed in a centrifuge. X-ray diffraction measurement was performed for a powdered sample prepared from a single crystal. We detected no extra peaks corresponding to the impurity phases in the diffraction profile, which was found to show the cubic structure with the lattice parameter $a = 14.00$ Å. This was in agreement with a previous result.[8)] Electron probe microanalysis (EPMA) confirmed the stoichiometric proportion of the crystal. The electrical resistivity was measured along the [100] direction by a standard four-probe ac method using

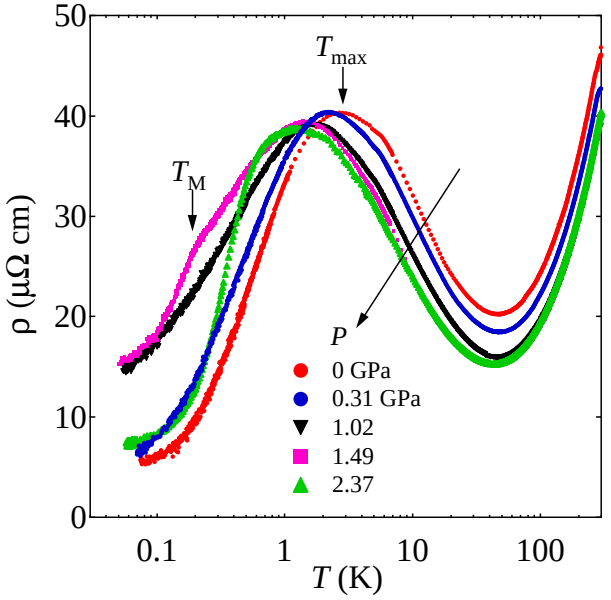

Fig. 1. (Color online) Electrical resistivity of $YbCo_2Zn_{20}$ as a function of the temperature. T_{max} and T_M correspond to the resistivity maximum temperature and magnetically ordered temperature, respectively. (see text for details).

an ac resistancebridge (model LR700, Linear Research Inc.). A single crystal sample with a typical dimension of $1.2 \times 0.5 \times 0.2\,\mathrm{mm}^3$ was prepared by polishing. The sample was cooled to 50 mK using a ^{3}He/^{4}He dilution refrigerator. Hydrostatic pressure was generated by using a hybrid-piston-cylinder-type pressure cell and Daphne oil 7373 as the pressure-transmitting medium. The hybrid pressure cell comprised of an inner shell and an outer jacket was made of NiCrAl and hardened CuBe alloys.[10] The pressure at low temperatures was determined by the superconducting transition temperature of a small piece of lead placed inside the pressure cell.[11]

Figure 1 shows the temperature dependence of the electrical resistivity $\rho(T)$ at selected pressures. The sample used in the present experiment exhibits a smaller residual resistivity ($\rho_0 \sim 5\ \mu\Omega\,\mathrm{cm}$) than that of a previous report ($\rho_0 \sim 21\ \mu\Omega\,\mathrm{cm}$).[8] At the ambient pressure, $\rho(T)$ exhibits a minimum at ∼50 K. Below this temperature, $\rho(T)$ varies roughly as $-\ln T$ and shows a broad maximum at $T_{max} \sim 2.6$ K. Applying pressure does not change the overall temperature dependence of the resistivity, while T_{max} shifts to a lower temperature with an increase in the pressure.

In Fig. 2, we focus on the low-temperature part of the electrical resistivity at selected pressures. As shown in Fig. 2(a), $\rho(T)$ below T_{max} decreases with a decrease in the temperature. The application of pressure causes a change in the $\rho(T)$ behavior (see below for details); however, there is no pronounced anomaly indicating the magnetic ordering or superconducting transition down to 50 mK at low pressures ($P \leq 0.92$ GPa). When the pressure exceeds a critical pressure (∼ 1.02 GPa), the resistivity shows a kink at T_M (see the arrows in Fig. 2(b)) and T_M increases with an increase in the pressure. Here, T_M is defined as the temperature where $\frac{d^2\rho}{dT^2}$ shows

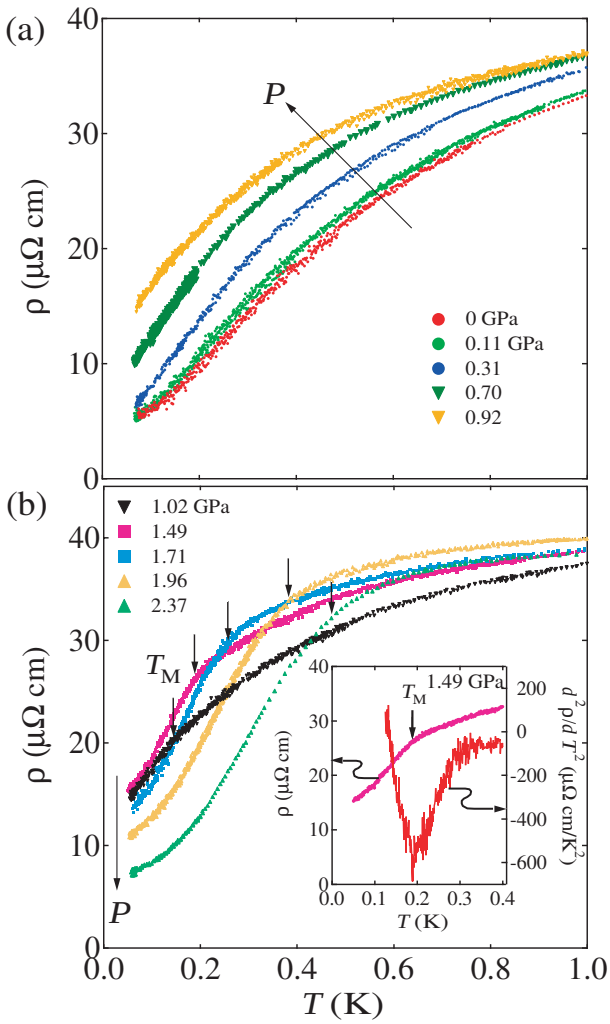

Fig. 2. (Color online) Temperature dependence of the electrical resistivity at low temperatures: (a) below and (b) above P_c. (b) The arrows indicate T_M. The inset shows $\rho(T)$ and $\frac{d^2\rho}{dT^2}$ as functions of the temperature at 1.49 GPa. T_M was estimated from the temperature where $\frac{d^2\rho}{dT^2}$ shows a minimum.

a minimum (see the inset of Fig. 2(b)).

In order to demonstrate the effect of pressure on the Fermi-liquid behavior described by $\rho(T) = \rho_0 + AT^2$, $\rho(T)$ is plotted as a function of T^2 in Fig. 3. Here, the coefficient A of the T^2 term of the resistivity is deduced from the slope in the linear portion of the plot. At the ambient pressure, $\rho\,(T)$ shows typical Fermi-liquid behavior at $T < T_{FL}$ (∼0.22 K), which gives the onset temperature of the T^2 dependence. With an increase in the pressure for $P < 0.6$ GPa, the slope of the solid line increases due to the strong enhancement of the A coefficient, while T_{FL} gradually decreases. At higher pressures between 0.70 and 1.34 GPa, the resistivity deviates from the T^2 dependence and appears to show T^n dependence for $1 \leq n < 2$.[12,13] Upon further increasing the pressure, the Fermi-liquid behavior is recovered.

Figure 4 shows the pressure dependence of A and ρ_0 for $YbCo_2Zn_{20}$. Note that we estimate ρ_0 by linear extrapolation to 0 K in the pressure range between 0.70 and 1.34 GPa where $\rho(T)$ does not obey the Fermi-liquid

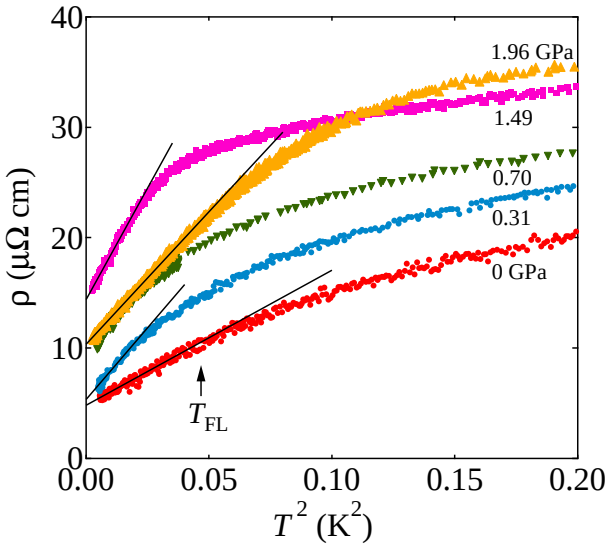

Fig. 3. (Color online) Electrical resistivity as a function of T^2 at selected pressures. The solid lines are fits to the functional form $\rho(T) = \rho_0 + AT^2$. T_{FL} corresponds to the upper limit of the T^2 dependence of the resistivity, as indicated by the arrow at the ambient pressure.

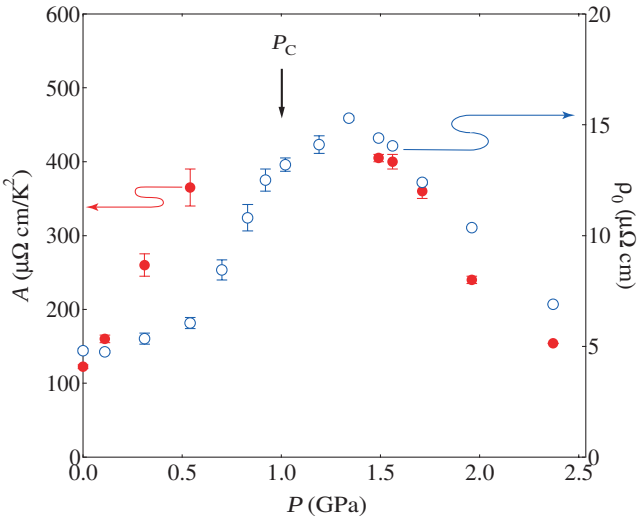

Fig. 4. (Color online) Pressure dependence of A and ρ_0.

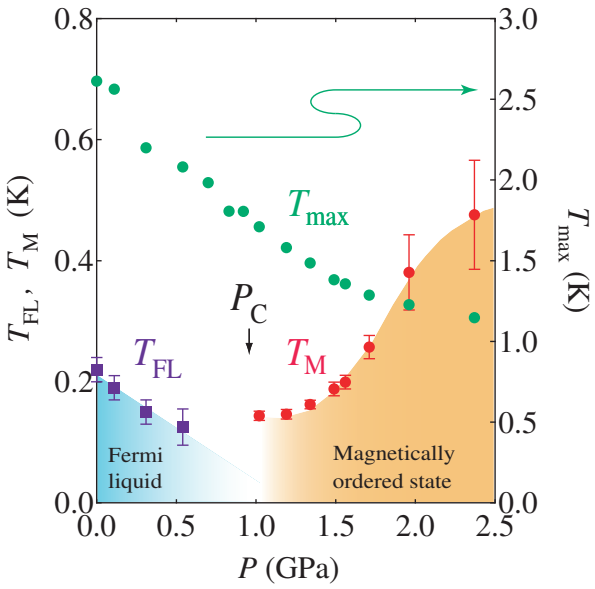

Fig. 5. (Color online) Pressure-temperature phase diagram of $YbCo_2Zn_{20}$.

behavior down to 50 mK. As the pressure approaches the critical pressure P_c, A monotonically increases to approximately 400 $\mu\Omega$ cm/K^2. The parameter ρ_0 also increases with the pressure and has a broad maximum slightly above P_c. As is often the case with heavy fermion systems, critical fluctuations lead to the enhancement of A coefficient and ρ_0 in the vicinity of a magnetic QCP,[14] suggesting the appearance of the magnetically ordered phase above P_c. Consequently, we speculate that the resistivity anomaly at T_M is due to the appearance of the magnetically ordered state at low temperatures. The anomalous temperature dependence of the resistivity in the vicinity of P_c is interpreted in terms of the quantum critical behavior.[15]

In Fig. 5, we summarize the results for $YbCo_2Zn_{20}$ on a pressure-temperature phase diagram; the crossover temperature T_{FL} denotes the upper limit of the observed T^2 behavior in the resistivity, T_M is the temperature where the resistivity anomaly is detected, and T_{max} is the resistivity maximum temperature. T_{max} and T_{FL} are suppressed by applying pressure, and thus, the nonmagnetic Fermi-liquid state vanishes around P_c. T_M appears to abruptly develop above P_c, where T_{max} decreases. Assuming that the observed T_{max} in the resistivity is proportional to T_K,[16,17] the value of T_K decreases with an increase in the pressure. According to a theoretical study, the suppression of valence fluctuations leads to a decrease in T_K.[18] It is generally accepted that the magnetic instability results from the competition between the RKKY and the Kondo interactions. For $YbCo_2Zn_{20}$, the RKKY interaction becomes more dominant with a reduction in the Kondo interaction. As a result, we conclude that the magnetic ordering transition occurs at T_M above P_c. It is worth noting that P_c ($\sim$ 1 GPa) is considerably lower than that of other Yb compounds,[3–7] which is an advantage in investigating quantum critical phenomena. We believe that the relatively smaller P_c in $YbCo_2Zn_{20}$ is simply due to its proximity to the magnetic QCP at the ambient pressure, as inferred from the small T_K. T_M increases slightly with an increase in the pressure up to $\sim$1.4 GPa and subsequently increases rapidly at higher pressures. It is interesting that ρ_0 exhibits a maximum at $\sim$ 1.4 GPa. Similar behavior has also been observed in other Yb compounds showing pressure-induced magnetic ordering, e.g., $YbCu_2Si_2$ and $YbNi_2Ge_2$.[4,5] In the case of $YbCu_2Si_2$, the magnetic moment of Yb ions is partially ordered at P_c and the maximum ρ_0 deep in the magnetic phase may coincide with the full ordering at higher pressures.[4] We believe that for $YbCo_2Zn_{20}$, this problem deserves further investigation.

Finally, we consider the crystalline electric field (CEF) effect in $YbCo_2Zn_{20}$. In a cubic symmetry environment, the eightfold degenerate ground state of the Yb^{3+} ion

($J = 7/2$) is split into two doublets and one quartet. In the grand Kadowaki-Woods relation,[19] $YbCo_2Zn_{20}$ is located near the line $N = 4$, where N is the degeneracy number of quasi-particles forming a heavy fermion system.[8] The transport and thermodynamic properties exhibit anomalous features due to the competition between the energy scale of the CEF splitting, Δ_{CEF}, and T_K. As observed in $YbNi_2Ge_2$,[5] the resistivity maximum is split into two maxima under pressure because the pressure drives the system from a state where $T_K > \Delta_{CEF}$ to one where $T_K < \Delta_{CEF}$. On the other hand, under pressures, $\rho(T)$ of $YbCo_2Zn_{20}$ continuously shows the single maximum. This result may be interpreted as indicating that the system is dominated by the Kondo interaction over the CEF splitting in the pressure range of this study. We infer that near spherical coordination of the Zn atoms surrounding Yb ions causes the CEF splitting to a relatively small extent.

In summary, we have measured the electrical resistivity of $YbCo_2Zn_{20}$ under pressure. Above a critical pressure P_c (~ 1 GPa), we have found a resistivity anomaly at T_M that increases with the pressure. The resistivity at low temperatures deviates from the T^2 behavior around P_c, while the system shows a nonmagnetic ground state described by the Fermi-liquid model below 0.6 GPa. The coefficient of the electrical resistivity A strongly increases close to P_c. From these results, we suggest that the magnetically ordered transition occurs above P_c in $YbCo_2Zn_{20}$.

To elucidate the pressure-induced magnetically ordered phase, further experiments are needed. In our recent experiments, the magnetic transition has been confirmed to be of bulk origin through specific heat measurements.[20] We believe that the present study deserves further investigation to obtain a deeper insight into the magnetic instability in heavy fermion compounds.

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