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Pressure effect on valence fluctuation and magnetic ordering in YbPd

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ABSTRACT

The cubic CsCl type compound YbPd is known to be a valence fluctuating compound and to undergo four phase transitions at 0.5, 1.9, 105 and 125 K. In the present study, we focus on pressure effect on the transitions. The transitions at $T_1 = 105$ K and $T_2 = 125$ K, which are accompanied by thermal hysteresis, are of first order. With increasing pressure, the transitions shift toward lower temperature direction. The transition at T_1 satisfies the Clausius–Clapeyron's relation, which also supports the first-order transition. T_1 disappears at around 1.5 GPa. Above 1.5 GPa, some new ground state would be realized. We can observe an anomaly at 2.8 K which would be associated with magnetic ordering at 1.9 K. The anomaly becomes more distinct with pressure. Since applying pressure stabilizes magnetic Yb^{3+}, the magnetic ordering becomes more conspicuous.

1. Introduction

The competition between Kondo effect and Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction in rare-earth compounds induces such exotic phenomena as valence fluctuation, heavy fermion [1], Kondo insulator [2], anisotropic superconductivity [3] and so on. Since these phenomena, which are ascribed to unstable 4f electrons, are quite sensitive to pressure, pressure is a powerful tool for investigation of the unstable behavior in rare-earth compounds.

The cubic CsCl type compound YbPd is known to be a valence fluctuating compound and to undergo four phase transitions at 0.5, 1.9, 105 and 125 K [4]. These transitions are confirmed by measurements of specific heat, electrical resistivity, ac susceptibility and thermal expansion [4]. The ^{170}Yb Mössbauer studies reveal that the one at 1.9 K is due to magnetic ordering [5]. However, mechanisms of the other three phase transitions remain unknown. The X-ray absorption spectroscopy (XAS) at the Yb L_{III}-edge manifests Yb valence is ~2.8+ and almost independent of temperature even at 105 and 125 K [4]. This result means the phase transitions at 105 and 125 K are not a valence transition. The Mössbauer studies also suggest two different Yb charge states are present in equal proportions at the lowest temperature, which suggests charge ordering though Yb ion occupies only one crystallographic site in the CsCl type structure [5]. In spite of such interesting features, YbPd has not been examined for a long time. In the present study, we focus on pressure effect on the transitions. We have measured magnetic susceptibility electrical resistivity ρ in the temperature range between 2 and 300 K under high pressure.

2. Experimental methods

To prepare a polycrystalline sample of YbPd, an ingot of ytterbium metal and a powder of palladium metal with the purity of 99.9% and 99.95%, respectively, were used as a starting material. The mixture of shavings of the Yb metal and the Pd powder in a ratio of 1.3:1 was pressed into a pellet. The pellet, which was sealed into a tantalum tube under argon atmosphere by using an arc-furnace, was heated at 1450 °C for 40 h. Powder X-ray diffraction pattern displays the sample is in the cubic CsCl type structure with a lattice constant of $a = 3.439$ Å. As described in Ref. [5], a little broadening of the diffraction line was observed. Magnetization measurement under high pressure up to 1.0 GPa was carried out by using the magnetic properties measurement system (MPMS) manufactured by Quantum Design. Electrical resistivity was measured by ac four-probe method under high pressures up to 2.4 GPa in the temperature range from 2 K to room temperature. The pressure was generated by using piston cylinder type pressure cell and a 1:1 mixture of Fluorinert FC70 and FC77 as a pressure transmitting medium. The pressure cell for magnetization measurement is made of a non-magnetic CuTi alloy to reduce a magnetic signal of the pressure cell. The one for electrical resistivity measurement is a hybrid type cell which is composed of inner NiCrAl cylinder and outer CuBe one. To calibrate pressure, pressure dependence of superconducting transition temperature of a tin plate was examined.

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3. Results and discussion

Fig. 1 and its inset show temperature dependence of inverse magnetic susceptibility and magnetic susceptibility, respectively, at various pressures. For ambient pressure, the susceptibility exhibits no clear anomalies at 105 and 125 K. Above 150 K, the susceptibility obeys Curie–Weiss law with an effective Bohr magneton number of $\mu_{eff} = 3.69\mu_B$ and Weiss temperature of $\Theta_p = -95.5$ K, which are in good agreement with those reported in Ref. [6]. Compared with a theoretical μ_{eff} value of Yb^{3+} ($4.54\mu_B$), the experimental value is quite smaller, which would reflect the valence fluctuation of Yb ion. Assuming that the smaller experimental μ_{eff} value is ascribed to mixture of magnetic Yb^{3+} and nonmagnetic Yb^{2+}, we estimate the numerical ratio of $Yb^{3+} : Yb^{2+}$ to be 66:34 from the μ_{eff} ratio squared of $(3.69/4.54)^2$. The obtained ratio corresponds to Yb valence of 2.66+, which is comparably close to the Yb valence determined from the XAS (~2.8+). The large negative value of the Weiss temperature, Θ_p, is also peculiar to a valence fluctuating system. Therefore, the behavior of the magnetic susceptibility above 150 K seems to be associated with the valence fluctuation. Below 150 K, the susceptibility deviates upward from the Curie–Weiss law. Mechanism of the deviation, which seems to be associated with the transition at 105 and/or 125 K, is unknown. Actually, the temperature, where the susceptibility begins to deviate from the CW law, seems to shift toward lower temperature, which corresponds to the behavior of the electrical resistivity shown later. Under high pressure, the behavior of the susceptibility is qualitatively unchanged, which means the Yb ion remains valence fluctuating. With increasing pressure, the μ_{eff} value increases slightly and the Θ_p value, of which magnitude decreases, remains negative, which corresponds to increase in ratio of magnetic Yb^{3+} and decrease in Kondo temperature, respectively. These results corresponds to general trend that application of pressure stabilizes localized magnetic moment of Yb^{3+}.

Fig. 2 shows temperature dependence of the electrical resistivity ρ under various pressures. For ambient pressure, the resistivity decreases with decreasing temperature, which is metallic behavior. The shape of the $\rho-T$ curve is quite similar to that reported previously [4]. The curve has two steps accompanied by thermal hysteresis. The thermal hysteresis, which has not been reported in the previous study, is an evidence of a first-order phase transition. We estimated the phase transition temperature from the maximum of $d\rho/dT$, as shown in the inset of Fig. 2. There exist a definite peak and a broad shoulder at around 105 and 125 K, respectively, which correspond very well to the transitions reported by Pott et al. [4]. For convenience, we define the lower transition temperature and the higher one as T_1 and T_2, respectively. With increasing pressure, the transitions at T_1 and T_2 shift toward lower temperature, which is similar to the behavior of the susceptibility under high pressure shown in Fig. 1. Simultaneously, the shoulder at T_2 in the $d\rho/dT-T$ curve becomes more indistinct. Therefore, we give up examining pressure dependence of T_2. Now we are measuring thermal expansion under high pressure and can observe decrease in T_2 with pressure [7]. As shown in Fig. 3, T_1 decreases almost linearly with increasing pressure up to 1.25 GPa, and shows quite different pressure dependence at $P > 1.25$ GPa, where the thermal hysteresis is collapsed and thus the $d\rho/dT$ maximum probably denotes no longer the phase transition temperature. These results suggest that the transition at T_1 disappears at $P \geq 1.62$ GPa. Extrapolation of the data at $P \leq 1.25$ GPa to $T_1 = 0$ K gives us a critical pressure of ~1.5 GPa. For $P > 1.5$ GPa, the high-temperature phase ($T > T_1$) is stabilized down to zero temperature and some new ground state might be realized. For $P \geq 1.91$ GPa, the $\rho-T$ curve is almost independent of pressure.

From the data plotted in Fig. 3, the dT_1/dP value is found to be ~ -65 K/GPa. According to the results reported in Ref. [1], the values of $\Delta V/V$ and ΔS associated with the transition at T_1 are -0.3% and 1.2 J/K mol, respectively. Therefore, the

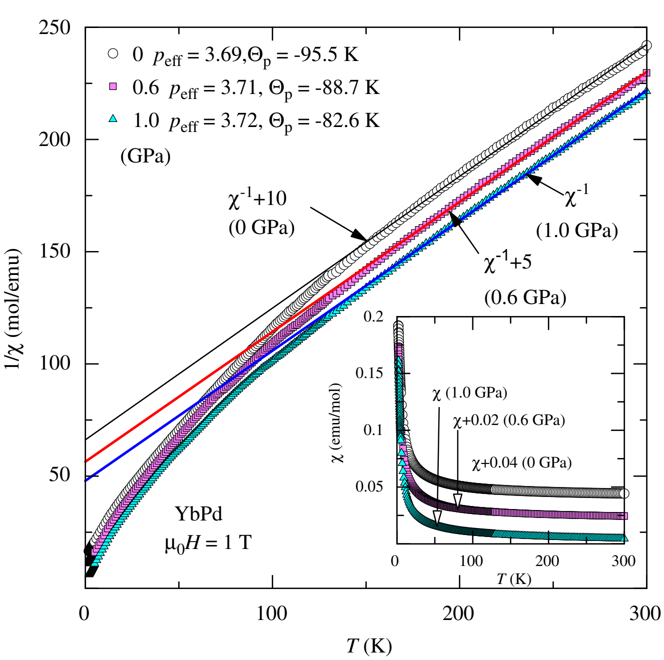

Fig. 1. Temperature dependence of inverse magnetic susceptibility of YbPd at various pressures. The solid line shows a Curie–Weiss law. The effective Bohr magneton number, μ_{eff}, and Weiss temperature, Θ_p, estimated from the Curie–Weiss law, are indicated for each pressure. The inset shows magnetic susceptibility as a function of temperature.

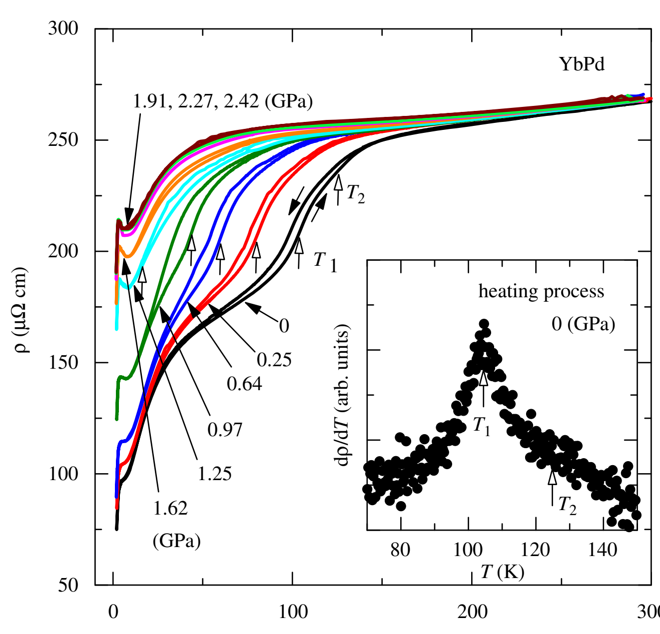

Fig. 2. Temperature dependence of the electrical resistivity of YbPd under high pressure. The white arrows show phase transition temperatures, T_1 and T_2. The inset exhibits $d\rho/dT$ as a function of temperature. We define T_1 as a peak position.

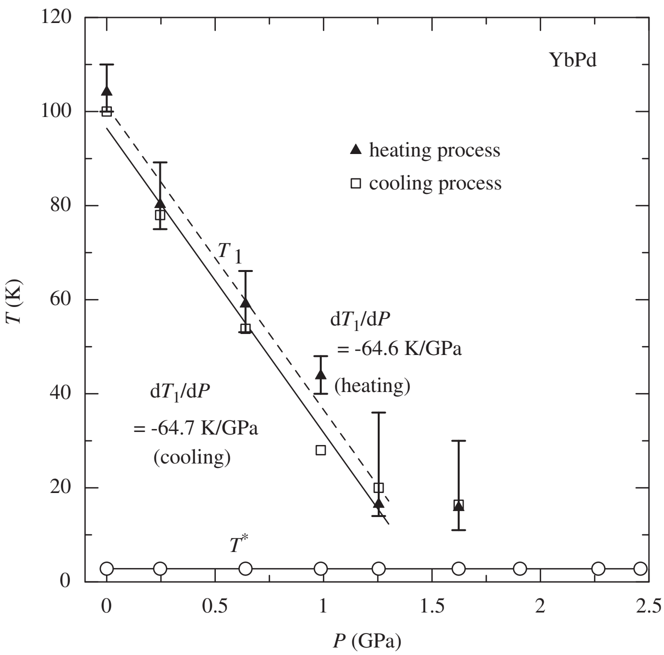

Fig. 3. Pressure dependence of the transition temperature T_1 measured during heating and cooling. The error bar is shown only for heating process. The rate of dT_1/dP is estimated to be ~ -65 K/GPa. The T^*, which would be associated with magnetic ordering, is also plotted as a function of pressure.

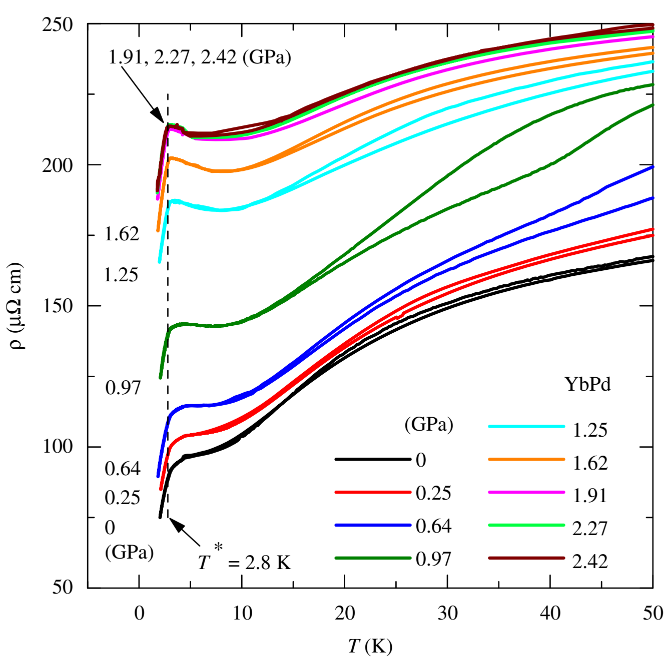

Fig. 4. Temperature dependence of the electrical resistivity of YbPd under high pressures in the temperature range between 2 and 50 K. At around 2.8 K, a bend or a peak is observed at all pressures studied.

Clausius–Clapeyron's relation, $dT/dP = \Delta V/\Delta S = -61$ K/GPa, is almost satisfied at the transition at T_1, which also supports the transition at T_1 is of first order.

Fig. 4 shows temperature dependence of the resistivity in the temperature range of 2–50 K. With increasing pressure, ρ value at the lowest temperature increases definitely. Especially, at $0.97 \leq P \leq 1.62$ GPa, where the transition at T_1 is being collapsed, the ρ value rises drastically, which is possibly associated with the high-temperature ($T > T_1$) phase stabilized down to zero temperature. In addition, a bend, which is observed at $T^* = 2.8$ K, becomes sharper and is transformed into a peak at $P \geq 1.25$ GPa. Namely, the anomaly, which remains at 2.8 K as shown in Fig. 3, becomes more evident with pressure. Below 2.8 K, the resistivity drops suddenly, which is reminiscent of disappearance of magnetic scattering. Since the application of pressure generally stabilizes magnetic Yb^{3+}, we speculate that the bend is associated with the magnetic ordering reported at 1.9 K [4] and that the magnetic ordering is more conspicuous. The $-\log T$-like behavior of ρ at around 5 K under higher pressures than 0.97 GPa reminds us of an impurity Kondo effect. Applying pressure would lower Kondo temperature, T_K, in Yb-based systems in contrast to Ce-based systems, which is also reported in cubic $YbCu_5$ [8,9].

The possibility of charge ordering at the lowest temperature has been pointed out by Bonville et al. [5] We speculate the transitions at T_1 and/or T_2, of which mechanism remains unknown, are associated with charge ordering. If so, the charge ordering would be collapsed under high pressure. Hereafter, we would like to verify the possibility.

4. Conclusions

In summary, we have reported the magnetic susceptibility and the electrical resistivity of YbPd under high pressure. We found the transitions at T_1 and T_2 are of first order. The transition at T_1 is collapsed at around 1.5 GPa. With applying pressure, an anomaly associated with magnetic ordering becomes more conspicuous, the resistivity at the lowest temperature is enhanced and $-\log T$ dependence of ρ appears. In the present study, we cannot investigate pressure dependence of T_2.

Acknowledgments

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