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High pressure studies of anomalous electronic states of $Y_{1-x}U_xPd_3$

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Abstract

The electrical resistivity $\rho(T)$ of $Y_{1-x}U_xPd_3$ ($x = 0, 0.05$ and 0.2) and the lattice constants for $x = 0$ have been measured at high pressure. It is found that the cubic Cu_3Au structure is stable up to 12 GPa at room temperature. The Kondo temperature T_K was extracted from the $\rho(T)$ curve and it was found that it increases with pressure. A logarithmic temperature dependence characteristic of the Kondo effect was found for $x = 0.2$ in the temperature range above about $0.5T_K$. Fermi liquid behaviour in $\rho(T)$ for $x = 0.05$, i.e., $\rho(T) \propto T^2$, is observed and its stability at high pressure is discussed. The pressure dependence of the Kondo temperature T_K is discussed using the Grüneisen parameters at T_K , Γ_K . It appears that the values of Γ_K are the same for these two compounds ($x = 0.05$ and 0.2): $\Gamma_K = 12$. The T -linear behaviour in $\rho(T)$ for $x = 0.2$, which is characteristic behaviour for non-Fermi liquids, is collapsed by an application of pressure and typical Fermi liquid quadratic temperature dependence recovers at high pressure. From the result for $x = 0.2$, the power n of the temperature in $\rho(T) \propto T^n$ is determined as 1.0 at ambient pressure and 1.9 at 5.8 GPa. It is pointed out that the hybridization effect due to the application of pressure gives rise to a crossover from a non-Fermi liquid state to Fermi liquid state. But the crossover temperature T_{cr} shows a pressure dependence different from that predicted by the two-channel Kondo model.

1. Introduction

It has been reported that some intermetallic compounds including U or Ce show a lot of interesting electronic properties such as heavy fermions (HF), the Kondo effect and

superconductivity [1]. Among them, the compounds showing behaviour substantially different from the normal Landau Fermi liquid (FL) type have been extensively studied [2–4]. The electronic state displaying these behaviours has been called the ‘non-Fermi liquid’ (NFL) state. As for the electrical resistivity, $\rho(T)$ for these compounds shows a weak power law at low temperatures, such as T^n ($n = 1–1.5$) [5, 6], which is in sharp contrast to that for normal FL behaviour: T^2 at low temperature. Anomalous behaviours have been observed for other physical quantities such as magnetic susceptibility and specific heat [3–5]. Several theoretical models from different points of view for interpreting these anomalous properties have been presented [4]. But the origin of the NFL state has not been completely understood yet.

The instability of the NFL state under the influence of perturbation or changing the control parameters has been studied extensively by many investigators [5–7]. In this area, high pressure study of NFL gives us a lot of important information because high pressure is considered as a ‘uniform perturbation’ and it gives rise to an electronic and structural phase transition or crossover in the materials having highly correlated marginal electronic states, which supplies a lot of crucial information for clarifying the electronic and magnetic structures of these materials [8]. Furthermore, since the disorder in the doped sample has been pointed out to play an important role in these systems [9, 10], high pressure study of undoped materials showing NFL behaviours is highly desired.

Until now, we have reported mainly, for several Ce compounds, on the pressure-induced crossover from the concentrated Kondo (CK) state or the HF state to the intermediate valence (IV) state in which the valence of the rare earth elements is not integer [11]. Since HF compounds generally have low Kondo temperature T_K but IV compounds have high T_K , the crossover is usually accompanied by a large increase of the Kondo temperature and a large decrease of the Grüneisen parameter for T_K at high pressure [12]. On the basis of high pressure work on the NFL, it has been reported that the NFL state becomes unstable at high pressure, showing a crossover to the normal FL [7, 13–16]. The results have been discussed on the basis of several different viewpoints. This also emphasizes the importance of pressure as a perturbation in tuning the electronic state of NFL compounds. However, in most work on pressure-induced crossover in NFL compounds (mostly for Ce compounds), almost all experimental research has been focused mainly on the ground state properties around 1 K or below, i.e., on the low temperature properties. But for Ce based compounds, we have pointed out the importance of the high temperature properties in connection with the Kondo effect [17].

$Y_{1-x}U_xPd_3$ exhibits a variety of interesting magnetic behaviours such as spin glasses, the Kondo effect and magnetic ordering, depending on x [3, 5, 18, 19]. Among these properties, the most interesting aspect of this pseudobinary compound is the NFL behaviours observed near $x = 0.2$: the approximately T -linear dependence ($n = 1.0–1.1$) in the electrical resistivity and a $T \log T$ dependence in the specific heat have been observed. In the high temperature range above about 80 K, a logarithmic temperature dependence in $\rho(T)$ was also observed, but there is no antiferromagnetic ordering at least down to 0.2 K [20]. This compound is a prototypical material showing NFL behaviour. With decreasing x , T_K increases, the NFL state quickly becomes unstable and then finally FL is stabilized at a low concentration of U ($x \leq 0.1$). As was mentioned in the foregoing section, T_K for CK and HF systems changes drastically on applying pressure. We expect also a lot of variety in the electronic states of $Y_{1-x}U_xPd_3$ compounds on tuning T_K by applying pressure. However, the crossovers from NFL to FL reported until now were mainly for doped samples at ambient pressure or limited to a relatively low pressure range below 2 GPa, which may not be enough to induce the crossover in the sample with $x = 0.2$. It is interesting to observe an electronic crossover of this prototypical NFL compound at high pressure above 2 GPa and to compare the electronic state with those of other materials showing NFL and/or FL behaviour.

In the present work we measured the temperature dependent electrical resistivity $\rho(T)$ for $Y_{1-x}U_xPd_3$ compounds having $x = 0.05$ and 0.2 at high pressure in order to examine the electronic crossover from the NFL to the FL state (and vice versa) induced by high pressure and furthermore the stability of the Kondo state under pressure. The lattice constants for $x = 0$ (YPd_3) are observed to obtain the bulk modulus and to check the pressure-induced crystal structure change. The present results are explained in connection with the $x-T$ phase diagram and the Grüneisen parameters for the Kondo temperature T_K . The electronic state for $x = 0.2$ at low temperature will be discussed, taking into account critical fluctuation near the phase boundary, and then compared with several theoretical models.

2. Experimental procedures

2.1. Sample preparation

The polycrystalline samples of $Y_{1-x}U_xPd_3$ with $x = 0, 0.05$ and 0.2 were prepared by arc melting the constituent elements under an argon atmosphere [18]. All the samples were remelted several times to ensure homogeneity and annealed in evacuated quartz tubes for 100 h at $900^\circ C$. X-ray diffraction analysis shows that all the samples are single phase with the cubic Cu_3Au type crystal structure. The lattice parameter a (\AA) changes linearly as a function of U concentration x as $a = 4.0701 + 0.0193x$ (\AA).

2.2. X-ray diffraction study under high pressure

Crystal structure and lattice constants under high pressure were investigated by means of x-ray diffraction. Hydrostatic pressure was generated by using WC Bridgman anvils having a face of 3 mm diameter [21]. A 4:1 methanol/ethanol mixture was used as a pressure transmitting medium. The powdered sample and NaCl were placed in a 0.3 mm hole at the centre of a beryllium disc gasket having 0.5 mm thickness. In order to obtain diffraction patterns with sharp lines and low background, we used a Guinier type focusing camera with a bent quartz monochromator. The diffraction lines were recorded on highly sensitive curved film. The pressure was determined by using Decker's equation of states for NaCl [22]. The $P-V$ relation was observed only for YPd_3 ($x = 0$) at room temperature.

2.3. Electrical resistance measurements under high pressure

Electrical resistance was measured in the temperature range between 4.2 and 300 K by using a standard dc four-probe method. Hydrostatic pressure below 2 GPa was generated by using a WC piston and a Cu-Be cylinder device. A 1:1 mixture of Fluorinert, FC70 and FC77, was used as a pressure transmitting medium. Above 2 GPa, we used a cubic anvil type high pressure apparatus. The pressure was changed only at room temperature in order to minimize the internal strain in the sample and kept constant within $\pm 1\%$ throughout each measurement. The details of the high pressure systems were reported previously [23, 24].

3. Results

3.1. Pressure dependence of the volume and electrical resistance at room temperature

Figure 1 shows the lattice constants of YPd_3 as a function of pressure below 12 GPa at room temperature. The cubic Cu_3Au crystal structure is stable up to 12 GPa since no new diffraction lines were observed at high pressure. It was found that the compression curve could be

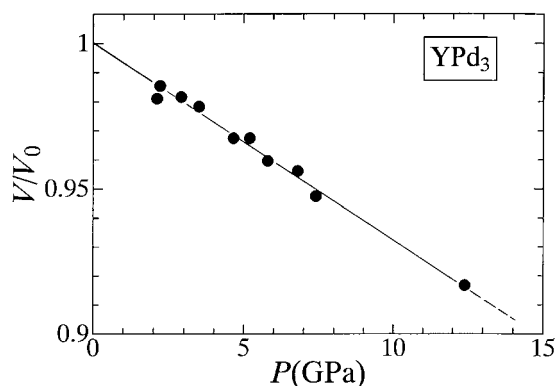


Figure 1. The pressure dependences of the lattice constants of YPd_3 at room temperature.

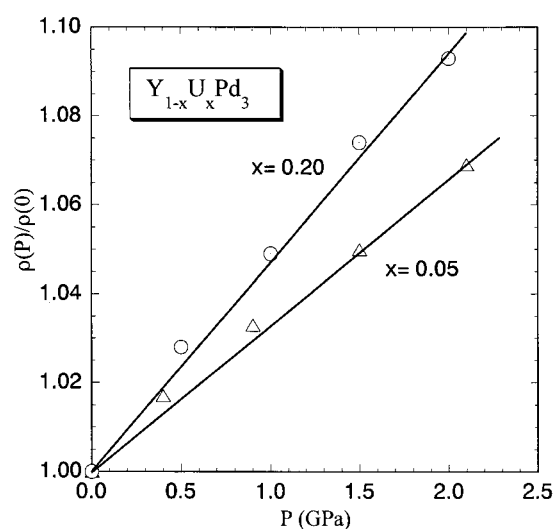


Figure 2. The relative change of the electrical resistivity, $\rho(P)/\rho(0)$, for $x = 0.05$ and 0.2 at room temperature.

approximated by a straight line, as shown in the figure. The bulk modulus B ($= -V\partial P/\partial V$) is estimated to be 148 GPa, which is smaller than that of URu_2Si_2 ($B = 215$ GPa) [25] but larger than that of UGe_2 ($B = 69$ GPa) [26]. Considering the fact that the lattice constant for $x = 0.2$ is almost the same as that for $x = 0$ [5], the bulk moduli for $x = 0, 0.05$ and 0.2 may be nearly the same. This result will be used in the following section when we calculate the Grüneisen parameters.

The normalized electrical resistivity $\rho(P)/\rho(0)$ for $x = 0.05$ and 0.2 at room temperature is shown in figure 2 as a function of pressure up to 2 GPa, where $\rho(0)$ is the resistivity at ambient pressure. ρ for the two samples increases with increasing pressure, approximately in a linear fashion. The pressure coefficient of ρ is estimated to be $(1/\rho)(\partial\rho/\partial P) = 30 \times 10^{-3} \text{ GPa}^{-1}$ for $x = 0.05$. The pressure coefficient of ρ for $x = 0.2$ is $46 \times 10^{-3} \text{ GPa}^{-1}$, which is 1.5 times larger than that for $x = 0.05$, suggesting that the conduction electrons are scattered in a complex way on the border of the instability of the electronic state near $x = 0.2$, which may give rise to an increase in the pressure coefficient of ρ . The electrical resistivity for $x = 0.2$

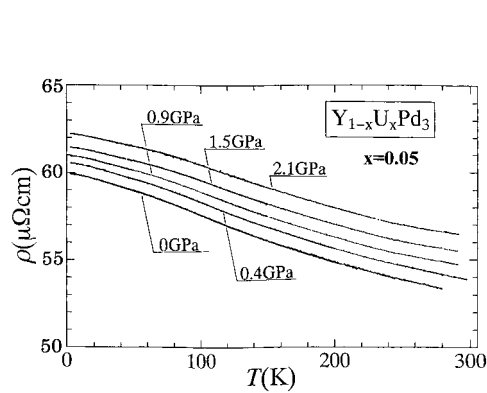


Figure 3. The electrical resistivity for $x = 0.05$ as a function of temperature at various pressures.

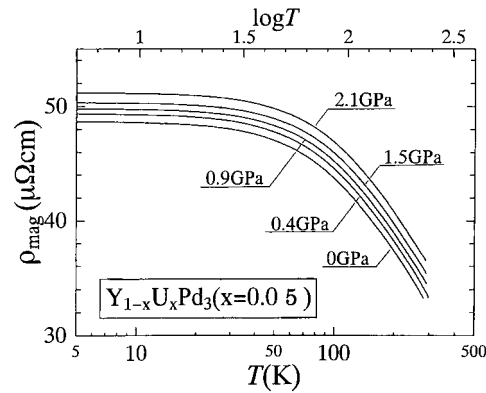


Figure 4. The electrical resistivity for $x = 0.05$ with a logarithmic scale for temperature at various pressures.

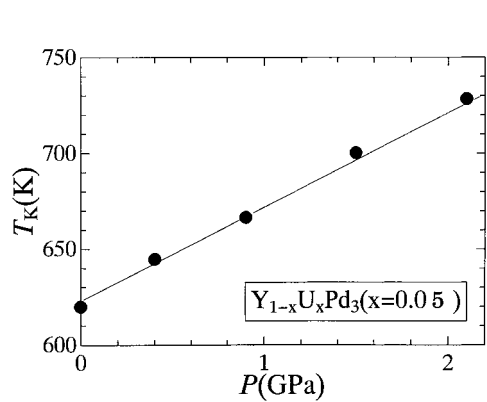


Figure 5. The Kondo temperature T_K for $x = 0.05$ as a function of pressure.

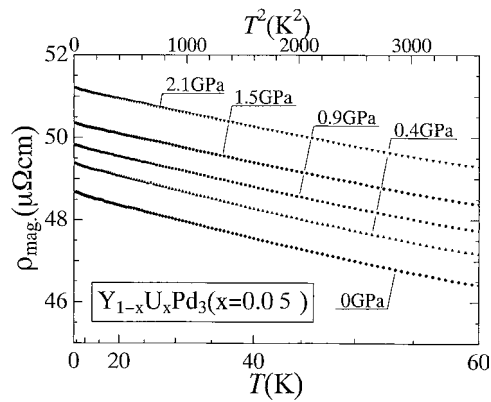


Figure 6. $\rho(T)$ as a function of T^2 for $x = 0.05$ at high pressure.

is found to increase with pressure up to 6 GPa at room temperature, having almost the same pressure coefficient as mentioned above.

3.2. Temperature dependent electrical resistivity for $x = 0.05$ under high pressure

Figure 3 shows the $\rho(T)$ curve at various pressures up to 2.1 GPa. $\rho(T)$ shows a smooth decrease with increasing temperature even at high pressure; all $\rho(T)$ curves seem to be almost parallel without any crossing but the values of ρ increase with pressure as was mentioned in relation to figure 2. The magnetic part of the resistivity ρ_{mag} was estimated by the equation $\rho_{\text{mag}}(T) = \rho(T, x = 0.05) - \rho(T, x = 0 (\text{YPd}_3))$, assuming that the $\rho(T)$ of YPd_3 (see figure 8 in the following section) is mainly dominated by phonon scattering. Figure 4 shows the results; $\rho_{\text{mag}}(T)$ is shown with a logarithmic scale for T at various pressures.

In the present work, we define the value of the Kondo temperature T_K as the temperature where the value of $\rho_{\text{mag}}(T)/\rho_{\text{mag}}(T = 4.2 \text{ K})$ is 0.5 [27]. T_K for $x = 0.05$ was obtained by an extrapolation of the present $\rho_{\text{mag}}(T)$ curve to high temperature. Figure 5 shows T_K as a function of pressure. T_K increases almost linearly with pressure up to 2 GPa having a rate of

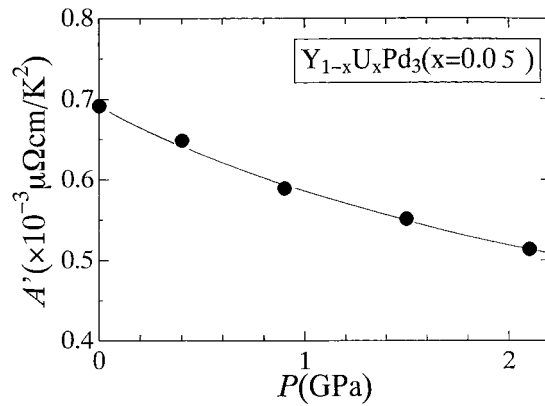


Figure 7. The coefficients of the T^2 term in $\rho(T)$ for $x = 0.05$, A' , as a function of pressure.

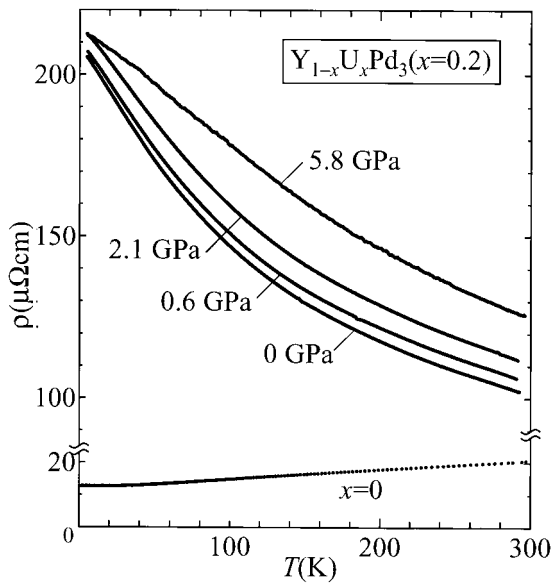


Figure 8. $\rho(T)$ as a function of T for $x = 0.2$ at high pressure including $\rho(T)$ for $x = 0$ as a reference.

$\partial T_K / \partial P = 50 \text{ K GPa}^{-1}$. Usually $\rho(T)$ for Kondo compounds shows normal Fermi liquid properties such as a T^2 dependence at low temperature. In order to examine this temperature dependence, we plotted the values of $\rho_{\text{mag}}(T)$ as a function of T^2 , in figure 6. A T^2 dependence in the $\rho_{\text{mag}}(T)$ curve, $\rho_{\text{mag}}(T) = \rho_0 - A'T^2$ (ρ_0 : residual resistivity), is found over the wide range of temperature below 50 K, which indicates that the ground state for $x = 0.05$ is well described as a Fermi liquid. The values of A' are plotted in figure 7 as a function of pressure. It is found that A' decreases with pressure: A' is decreased by about 30% on applying 2 GPa. Considering that A' is proportional to T_K^{-2} [28], this implies that T_K increases with pressure, which is consistent with the result in figure 5. We will discuss this point in detail later.

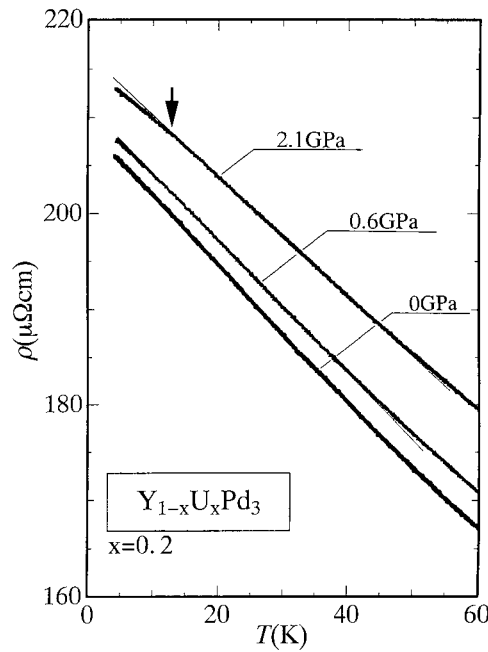


Figure 9. $\rho(T)$ for $x = 0.2$ below 60 K at various pressures. A small deviation from the linear temperature dependence is observed below 12 K at 2.1 GPa, which is shown by an arrow.

3.3. Temperature dependent electrical resistivity for $x = 0.2$ at high pressure

$\rho(T)$ for $x = 0.2$ is shown in figure 8 at various pressures together with that for $x = 0$ (YPd_3). It is evident that the $\rho(T)$ curve at 5.8 GPa is very different from those below 2 GPa, particularly below 100 K. Figure 9 shows the electrical resistivity below 60 K as a function of T at 0, 0.6 and 2.1 GPa. It is found that the linear temperature dependence in ρ , which is evidence of NFL behaviour, is well observed below 50 K; i.e., $\rho(T)$ below 50 K is approximated by the equation $\rho(T) = \rho_0(1 - T/T_1)$, where ρ_0 is the residual resistivity and T_1 is the characteristic temperature which may be related to the Kondo or spin fluctuation temperature. But at 2.1 GPa, a small deviation from the linear temperature dependence is found below about 12 K, which may be a precursor of the collapse of the NFL state. We will discuss later the origin of this deviation. Figure 10 shows the coefficients A of the T -linear term, $A = \rho_0/T_1$, and T_1 as a function of pressure. The magnitude of A at ambient pressure ($0.72 \mu\Omega \text{ cm K}^{-1}$) is nearly the same as those reported previously [4]. The value of A is found to decrease with increasing pressure, having a rate $(1/A)(\partial A/\partial P) = -5.7 \times 10^{-2} \text{ GPa}^{-1}$. From the linear extrapolation of $A(P)$, A is 0 around 18 GPa, where the NFL state is expected to disappear.

Figure 11 shows the magnetic part of the electrical resistivity $\rho_{\text{mag}}(T)$ at various pressures with a logarithmic scale for T , which was estimated by means of the same method as mentioned in section 3.2. The Kondo temperature T_K was also defined in the same way as that for $x = 0.05$. The results are shown in figure 12. T_K increases almost linearly with increasing pressure below 2 GPa, but the point at 5.8 GPa deviates upwards slightly from that extrapolated from low pressure below 2 GPa. It should be noted that the effect of pressure on T_K is opposite to the effect of the U concentration x [5, 18]: T_K decreases with x but increases with pressure.

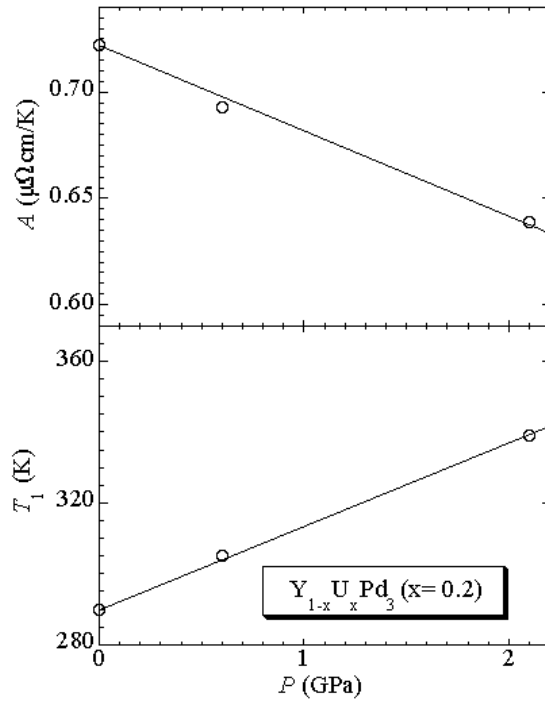


Figure 10. The coefficients of the linear term A and the characteristic temperature T_1 as a function of pressure for $x = 0.2$.

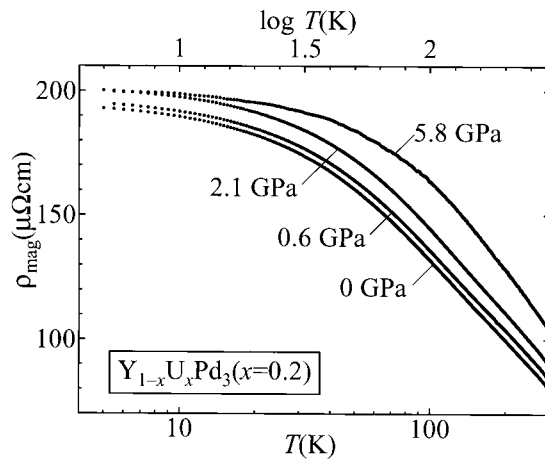


Figure 11. The magnetic part of the electrical resistivity $\rho_{\text{mag}}(T)$ for $x = 0.2$ on a logarithmic scale at high pressure.

4. Discussion

In this section we discuss the experimental results obtained in the present work in connection with the Kondo effect and the crossover from a NFL state to a FL one. We divide the discussion into four parts.

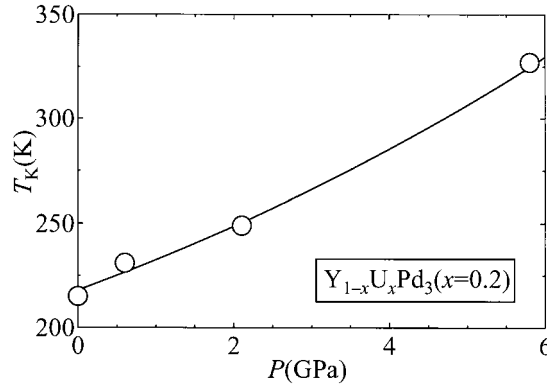


Figure 12. The Kondo temperature T_K for $x = 0.2$ as a function of pressure.

4.1. Stability of the Kondo state under high pressure—high temperature electronic properties above 100 K

We discuss the high temperature behaviour of $\rho(T)$, in which Kondo scattering dominates $\rho(T)$. In the foregoing section, we obtained the Kondo temperature T_K as a function of pressure, where we found an approximately linear dependence against pressure. T_K may be described by the following equation in the single-impurity regime:

$$T_K \propto \exp\left[-\frac{1}{|JN(0)|}\right], \quad (1)$$

where J is the s-f exchange interaction and $N(0)$ is the density of states at the Fermi level. J is written as

$$J \sim \frac{\langle V_{sf}^2 \rangle}{\epsilon_{5f}}, \quad (2)$$

where V_{sf} is the strength of hybridization and ϵ_{5f} is the energy difference between the 5f level and the Fermi level. ϵ_{5f} may be expressed as $\epsilon_{5f} \sim \epsilon_0 + \epsilon_1 x - \omega P$, where ϵ_0 , ϵ_1 and ω are constants [5, 29]. This indicates that the effect of x on J or T_K is opposite to that of pressure, which is the same as the case for CeR alloys (R: rare earth element) [29].

Since V_{sf} increases and ϵ_{5f} decreases with increasing pressure, J shows an increase on applying pressure. This indicates that T_K is enhanced by applying pressure. The pressure dependence of T_K is derived from equations (1) and (2) as $T_K \sim (T_K)_0 e^{\gamma P}$, where $(T_K)_0$ is T_K at $P = 0$ and γ is a positive constant. In this derivation, we assumed that $\langle V_{sf}^2 \rangle$ and $N(0)$ are constant and pressure is not too high. The solid line in figure 12 was plotted by assuming $\gamma = 7.3 \times 10^{-2} \text{ GPa}^{-1}$ and $T_K(0) = 215 \text{ K}$; the experimental results are well reproduced by this equation. On the same assumptions, when pressure is constant, we can derive the following relation for the concentration (x) dependence of T_K : $T_K = T_K(0) \exp(-\theta x)$, where θ is a positive constant. This means that $T_K(x)$ decreases with increasing U concentration.

On the other hand, the value of γ is obtained as $8.0 \times 10^{-2} \text{ GPa}^{-1}$ for $x = 0.05$ by using the data for T_K shown in figure 5. These two values are approximately the same. This means that as far as the effect of pressure on the Kondo state is concerned, there is no significant difference between $x = 0.05$ and 0.2. In other words, the high temperature behaviour of $\rho(T)$ can be treated in the framework of the Kondo single-impurity model.

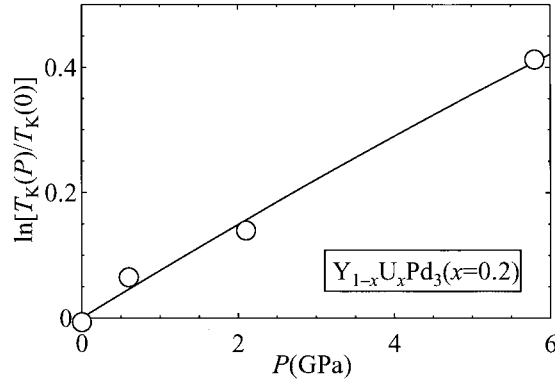


Figure 13. $\ln[T_K(P)/T_K(0)]$ as a function of pressure for $x = 0.2$.

4.2. Estimation of Grüneisen parameters for $Y_{1-x}U_xPd_3$

The Grüneisen parameter is often used to evaluate the electronic state of highly correlated electron systems [8, 11, 12, 17, 25]. So we attempt to estimate this parameter for T_K . The Grüneisen parameter of T_K is defined as

$$\Gamma_K \equiv -\frac{\partial \ln T_K}{\partial \ln V} = B \frac{\partial \ln T_K}{\partial P}, \quad (3)$$

where B is the isothermal bulk modulus. The pressure dependences of T_K were shown in figures 5 and 12 for $x = 0.05$ and 0.2 , respectively. Equation (3) is rewritten as $\Gamma_K = B \partial \ln(T_K(P)/T_K(0))/\partial P$, where $T_K(0)$ is T_K at ambient pressure. Figure 13 shows the pressure dependence of the value of $\ln[T_K(P)/T_K(0)]$ for $x = 0.2$. The slope of the plot corresponds to Γ_K/B [12]. The value of Γ_K/B is estimated to be 0.08 GPa^{-1} . Γ_K for $x = 0.2$ is 12 at ambient pressure, assuming the value of B to be the same as that of YPd_3 ($=148 \text{ GPa}$), which is smaller than those of UAl_2 ($\Gamma \sim 20$) [8] and URu_2Si_2 ($\Gamma \sim 30$) [25]. It is interesting to note that the magnitude of Γ_K for $x = 0.2$ is comparable with that for the intermediate valence compounds $CePd_3$ ($\Gamma \sim 6$) [30] and $CeNi$ ($\Gamma \sim 9$) [31]. Γ_K for $x = 0.05$ is calculated in the same way, using the data in figure 5, to be 12, which is the same as that for $x = 0.2$. This result indicates that there is no significant difference in pressure effect on the Kondo state above 100 K between the $x = 0.05$ and 0.2 cases, as was pointed out in the foregoing section.

Here we apply the compressible Kondo model [32] to equation (1). The volume change of $|JN(0)|$ is assumed as

$$|JN(0)| = |JN(0)|_0 \exp\left(-q \frac{V - V_0}{V_0}\right), \quad (4)$$

where q is a constant between 6 and 8 and $|JN(0)|_0$ is $|JN(0)|$ at ambient pressure. By using equations (1) and (4) we obtain

$$|JN(0)|_0 = \frac{q}{\Gamma_K}. \quad (5)$$

When we derived equation (5), we used the following approximation: $\exp(q(V - V_0)/V_0) = 1 + q(V - V_0)/V_0$, which is valid at low pressure because the volume change is small. Assuming $q = 6$ [32], we obtain 0.5 for both $x = 0.2$ and 0.05 . This is larger than those for URu_2Si_2 (0.2) and UAl_2 (0.3), but the same as that for $\alpha\text{-Ce}$ (~ 0.5) [33]. The results indicate that the 5f electrons for $x = 0.2$ and 0.05 are more itinerant than those in URu_2Si_2 and UAl_2 .

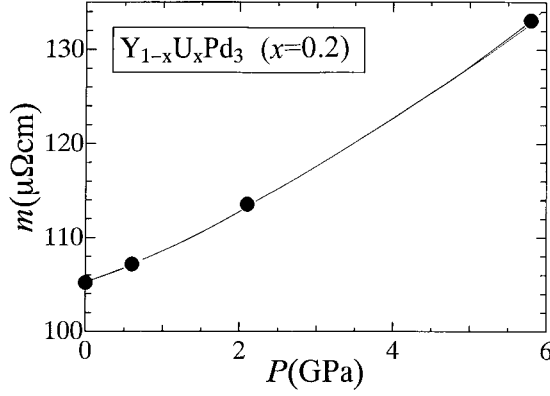


Figure 14. The coefficients of the $\log T$ term m for $x = 0.2$ as a function of pressure.

It is well known that the Kondo effect is manifested in the existence of a $\log T$ term in the electrical resistivity. The electrical resistivity of dilute Kondo alloys may be described as

$$\rho(T) = \rho_B - C|JN(0)|^3 \log T, \quad (6)$$

where C is the constant and ρ_B is the temperature independent resistivity calculated from Born first-order perturbation of J . From the above equation (6), the slope m in the ρ versus $\log T$ curve is derived to be

$$m = C|JN(0)|^3. \quad (7)$$

The values of m for $x = 0.2$ are plotted in figure 14 as a function of pressure. In this calculation of m , we used the data in the range $0.5T_K < T < 1.1T_K$ ($T_K = 215$ K at ambient pressure for $x = 0.2$). For this reason, we did not calculate the values of m for $x = 0.05$ because T_K for $x = 0.05$ is 620 K at ambient pressure and then $0.5T_K$ is 310 K, which is outside the temperature range in the present experiment. So in this section we limited our discussion to the $x = 0.2$ case. It is found that the value of m increases substantially with pressure for $x = 0.2$, as has been observed in many Ce based HF systems [12].

On the basis of the compressible Kondo model, we can explain the results in figure 14 as follows. $|JN(0)|^3$ is described from equation (4) as

$$|JN(0)|^3 \approx |JN(0)|_0^3 \left(1 + 3q \frac{V_0 - V}{V_0} \right). \quad (8)$$

From equations (7) and (8), we get the magnitude of m as proportional to $1 + 3q(V_0 - V)/V_0 \approx 1 + 3q\kappa P$, where κ is the compressibility. This implies that m increases with pressure, which explains qualitatively the results in figure 14.

4.3. Pressure-induced crossover from the NFL to the FL—low temperature electronic properties

Here we discuss first the relation between the values of T_K and the coefficients of the T^2 term for $x = 0.05$. In the Kondo compounds, the electrical resistivity at low temperature ($T \ll T_K$) is approximated by the following temperature dependence:

$$\rho(T) = \rho_0 \left(1 - \left(\frac{T}{T_K} \right)^2 \right), \quad (9)$$

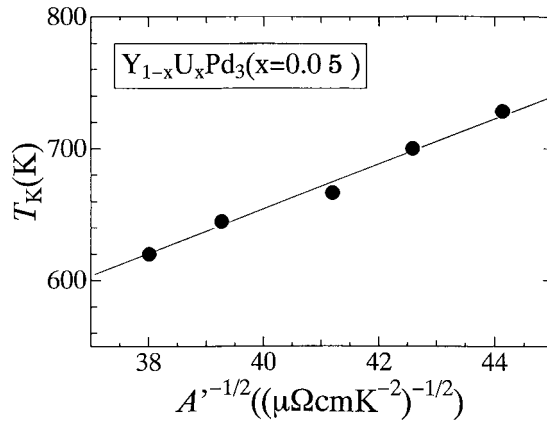


Figure 15. T_K as a function of $1/\sqrt{A'}$ for $x = 0.05$.

where ρ_0 is the residual resistivity. In this approximation, the value of A' is roughly proportional to $1/T_K^2$. Figure 15 shows the relation between T_K and $1/\sqrt{A'}$ at different pressures for $x = 0.05$. It is found that there is a good linear relationship between T_K and $1/\sqrt{A'}$. Using equation (9), we obtain the value of Γ_K as 13, which is in good agreement with the value obtained in section 4.2: 12. This suggests that the behaviour for $x = 0.05$ is well described by the single-impurity Kondo model over a wide range of temperature.

Next we consider the ground state for $x = 0.2$ at high pressure. The NFL behaviour below 2.1 GPa is evidenced by the existence of T -linear behaviour in $\rho(T)$, which was also observed below about 50 K at ambient pressure in the present work, as shown in figure 9. The coefficient A of the T -linear term decreases with increasing pressure, which may be explained by the increase of T_1 at high pressure. T_1 is related to some kind of fluctuation such as a spin or spatial one. Critical fluctuations associated with antiferromagnetic ordering were observed for $x = 0.2$ at low temperature down to 0.2 K using neutron scattering [20]. In this sense, the decrease of A (or the increase in T_1) suggests that the critical fluctuations near the phase boundary ($x \sim 0.2$) between the NFL state and spin glass state [5] are suppressed by application of pressure. If antiferromagnetic ordering took place, T_1 would become 0 and A would diverge near a critical value of a control parameter.

To allow comparison of the electronic state at 5.8 GPa and that at ambient pressure, the $\rho_{\text{mag}}(T)$ curves are shown in figure 16 along with that for $x = 0.05$, which shows the typical FL behaviour. It is easily seen that the overall behaviour of $\rho_{\text{mag}}(T)$ at 5.8 GPa is similar to that for $x = 0.05$ at ambient pressure. This implies that the electronic state for $x = 0.2$ at 5.8 GPa can be considered as a normal FL one.

Figure 17 shows the normalized resistivity $\rho_{\text{mag}}(T)/\rho_{\text{mag}}(4.2 \text{ K})$ as a function of the reduced temperature T/T_K at various pressures. The data below 2.1 GPa are found to fall on a universal curve. This indicates that the electronic state or NFL state is almost independent of pressure up to 2.1 GPa. But the curve at 5.8 GPa substantially deviates from the universal curve below 2.1 GPa. The large change in the temperature dependence suggests a crossover in the electronic state from the NFL to another state on applying pressure.

In order to see the result more clearly and examine the power law in T , we plot the values of $1 - \rho_{\text{mag}}(T)/\rho_{\text{mag}}(0)$ below 30 K in figure 18 both at ambient pressure and at 5.8 GPa as a function of T on a logarithmic scale. It is found that $\rho_{\text{mag}}(T)$ at low temperature below 30 K changes from a T -linear dependence ($n = 1.0$) at ambient pressure to a T^n ($n = 1.9$)

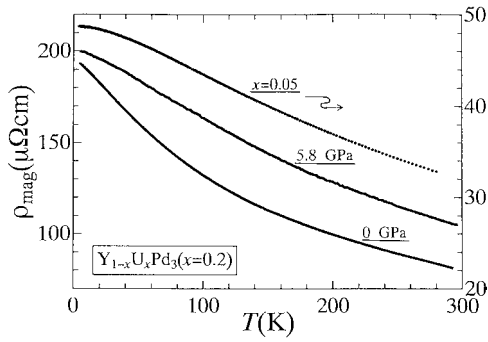


Figure 16. The temperature dependence of $\rho_{\text{mag}}(T)$ for $x = 0.2$ at 0 and 5.8 GPa and that for $x = 0.05$ for comparison.

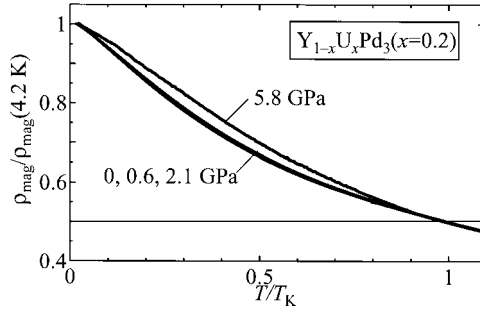


Figure 17. The normalized electrical resistivity, $\rho_{\text{mag}}(T)/\rho_{\text{mag}}(4.2 \text{ K})$, as a function of the reduced temperature, T/T_K , for $x = 0.2$.

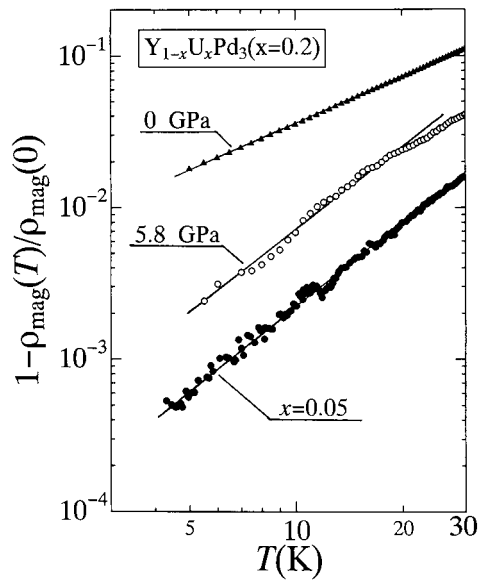


Figure 18. The magnetic part of the electrical resistivity for $x = 0.2$, $\rho_{\text{mag}}(T)$, as a function of $\log T$. The data for $x = 0.05$ are also shown for comparison.

dependence below about 17 K and the line for $x = 0.05$, which shows FL behaviour, is almost parallel to that at 5.8 GPa.

The change in the power from 1 to 1.9 suggests a collapse of the NFL state followed by the recovery of the FL state at high pressure. This result indicates that the crossover from NFL to FL states takes place at a pressure between 2.1 and 5.8 GPa, which is smaller than 18 GPa, where the value of A may vanish as was mentioned before. This is the first report of such a crossover induced by pressure for the prototype NFL compound $x = 0.2$.

Several theoretical models based on different viewpoints, for example the two-channel Kondo effect [34, 35], proximity to a quantum critical point [3, 36] and distribution of Kondo temperatures [37], have been proposed to explain the transport and thermodynamic properties of NFL compounds. However, there have been no theories succeeding in explaining all aspects of NFL behaviours [4]. Furthermore, it is not clear at present which model should be applied to explain the high pressure behaviour of NFL compounds. It is well known that the strength

of hybridization is increased by application of pressure and the electronic properties of the Kondo compounds have been interpreted in terms of a change in the hybridization caused by doping or pressure.

The effect of hybridization on the stability of FL and/or NFL compounds has been investigated by Koga and Shiba [38, 39] on the basis of the two-channel Kondo model. They calculated the phase diagram of the stability region of the FL and NFL as a function of Coulomb U and Hund couplings J_H .

According to their calculation, the enhancement of the strength of hybridization is found to stabilize the FL state compared with the NFL state. In other words, the application of pressure, which implies an increase of hybridization, gives rise to a crossover from NFL to FL behaviour. The present result seems to be explained qualitatively by taking their result into account as far as the crossover from the NFL to the FL state is concerned. Furthermore, the theory predicts that the temperature of crossover from NFL to FL behaviour, T_{cr} , decreases as the electronic state of the system approaches the NFL/FL boundary. T_{cr} may be described approximately as $T_{cr} \propto |J_H - J_{H,b}|^\alpha$, where $J_{H,b}$ is the value of J_H at the boundary and α is a constant, in the range 8–9 [40]. If we assume $T_{cr} = T_1$, the present result indicates that T_{cr} increases with increasing pressure as shown in figure 10, i.e., T_{cr} increases as the system approaches the FL/NFL boundary, which is in sharp contrast to the theoretical prediction. In other words, the present result suggests that there is no such boundary in the pressure change of the electronic state for $x = 0.2$ and the two-channel Kondo model may not be applicable in explaining the pressure change of characteristic temperatures or the pressure-induced crossover for $x = 0.2$.

The electrical resistivity $\rho(T)$ for NFL compounds has been calculated by Moriya and Takimoto on the basis of the self-consistent renormalization (SCR) theory of spin fluctuations [41]. Around the critical boundary, $\rho(T)$ shows a $T^{3/2}$ dependence followed by a T -linear dependence as T increases. Off the critical boundary region, $\rho(T)$ is proportional to T^2 , indicating the recovery of FL behaviour. As was shown in figure 18, $\rho(T)$ for $x = 0.2$ at 5.8 GPa is similar to that for $x = 0.05$, the Fermi liquid compound, which is also proved by the existence of a T^2 dependence at low temperature. The facts suggest that the spin fluctuation for $x = 0.2$ is expected to be suppressed by the application of pressure, which indicates an increase of T_1 , and then the NFL behaviour is collapsed to give rise to the recovery of the FL behaviour.

Recently, a phase diagram of heavy fermions including the quantum critical point has been presented by Continentino [42]; the Fermi liquid behaviour is expected to be stable off the critical region and the coherence temperature T_{coh} increases as the value of $|JN(0)|$ (or pressure) increases. If we assume T_{coh} as T_1 or T_K , the low temperature properties of $Y_{1-x}U_xPd_3$ ($x = 0.2$) may be qualitatively explained according to this diagram. In other words, since the sample for $x = 0.2$ is basically just on the quantum critical point, we expect the non-Fermi liquid behaviour and the pressure to have the effect of moving this sample off the critical point, which also gives rise to an increase in T_1 or T_K .

Roughly speaking, the common feature of these theories is that the NFL behaviours may collapse at high pressure showing a recovery of FL behaviour, but there is a difference between the semiquantitative explanations of the characteristic temperatures such as T_1 or T_K . Judging from the present results and the above-mentioned discussion, it is plausible to consider that the sample with $x = 0.2$ exists near or just on the quantum critical point [3, 36, 42] with a large magnetic fluctuation showing NFL behaviour, but with increasing pressure the quantum fluctuation is suppressed resulting in a crossover from the NFL to the FL state. In order to discuss which model is suitable for the pressure dependence of the NFL behaviour in the present work, more quantitative analysis of the experimental results is needed, not only for the resistance but also for the susceptibility or specific heat at high pressure. It seems difficult to explain all aspects of NFL behaviours by applying one theoretical model.

4.4. The relation between the U concentration x and the effect of pressure

In the compounds $Y_{1-x}U_xPd_3$, the lattice constants increase slightly with x , i.e., the lattice expands with x , which is opposite to the pressure effect. Here we derive the quantitative relation between the pressure P (GPa) and x by using the Kondo temperature T_K and lattice constant a as implicit variables. The fractional increase Δa (Å) in the lattice constant is obtained from the result in section 2.1 as a function of the U concentration x : $\Delta a = 19.3 \times 10^{-3} \Delta x$. On the other hand, the relation between the lattice constant and the pressure is obtained from the bulk modulus. From the bulk modulus in section 3.1, we obtain $\Delta a(P) = -9.2 \times 10^{-3} \Delta P$, where ΔP is in GPa. Using these two relations, we get the following equation:

$$\Delta P = -2.1 \Delta x. \quad (10)$$

Since the difference in x between $x = 0.05$ and 0.2 is -0.15 , the value of ΔP is estimated to be 0.3 GPa from the above equation (10). This result implies that the electronic state for $x = 0.2$ becomes similar to that for $x = 0.05$ on applying 0.3 GPa. But in section 4.3, we observed that the electronic state for $x = 0.2$ becomes the same as that for $x = 0.05$ above 4–5 GPa, which is extremely large compared to the estimated value—by an order of magnitude. This indicates that scaling using the lattice constants or the volume is impossible for this compound and the pressure-induced crossover from NFL to FL behaviour for $x = 0.2$ cannot be explained by just considering the lattice compression.

The Kondo temperature T_K of this material decreases with increasing x [5, 18]. Next we consider this using T_K as an implicit parameter. In section 4.1, we showed that T_K can be described as $T_K(P) = (T_K)_0 \exp(\gamma P)$. The fractional change in T_K is derived as $\Delta T_K/T_K = \gamma \Delta P$. On the other hand, $\ln T_K$ at ambient pressure is well known to be a linear function of x because of the relation $T_K(x) = T_K(x = 0) \exp(-\theta x)$ [5]. We get $\Delta T_K/T_K = -\theta \Delta x$. Thus we get the relation $\Delta P = -\theta/\gamma \Delta x$. By using the pressure dependence of T_K and $\theta = 7.1$, we obtain

$$\Delta P = -88 \Delta x. \quad (11)$$

If we put $\Delta x = -0.15$, we get $\Delta P = 13$ GPa. This means that about 13 GPa is needed for $x = 0.2$ to get the same electronic state as for $x = 0.05$. It is interesting to note that a pressure of several GPa is needed to destroy the NFL state for $x = 0.2$, which is comparable with the pressure mentioned above. This indicates that the change in the ‘effective pressure’, ΔP , is not due to just a change in the lattice constant but also a change in the electronic state followed by a small change in the lattice constant. Taking these results into account, the electronic state for $Y_{1-x}U_xPd_3$ compounds, particularly for $x = 0.2$, can be considered to be a marginal state on the border of magnetic instability.

5. Conclusion

In the present study, we have measured the electrical resistivity of $Y_{1-x}U_xPd_3$ ($x = 0, 0.05$ and 0.2) and lattice constant of YPd_3 at high pressure. The main conclusions are summarized as follows:

- (1) The Cu_3Au structure of YPd_3 is stable up to 12 GPa at room temperature.
- (2) As far as the pressure effect on the Kondo state above 100 K is concerned, there is no great difference between $x = 0.05$ and 0.2 because the Grüneisen parameter for T_K for $x = 0.2$ is the same as that for $x = 0.05$.
- (3) The non-Fermi liquid behaviour at ambient pressure for $x = 0.2$ shows a crossover at high pressure above 4–5 GPa, followed by a recovery of Fermi liquid behaviour.

- (4) The quantitative relation between pressure and U concentration has been discussed and the change in the internal (or effective) pressure is mainly dominated by the contribution from a change in the electronic state induced by a small compression in the lattice constant.

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