Pressure-dependent resistivity studies and the origin of non-Fermi-liquid behaviour in $U_xY_{1-x}Pd_3$

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Pressure-dependent resistivity studies and the origin of non-Fermi-liquid behaviour in $U_xY_{1-x}Pd_3$

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Abstract
We have studied the pressure and magnetic field dependence of the anti-ferromagnetic transition of $U_{0.45}Y_{0.55}Pd_3$. T_N increases with pressure whereas there is very little magnetic field dependence up to 7 T. These results indicate that $(U, Y)Pd_3$ is located in the weak-coupling regime of the Doniach phase diagram, contradicting some previous suggestions. From a comparative study of the resistivity of $U_{0.2}Y_{0.8}Pd_3$ under pressure and magnetic field, we have shown that the non-Fermi-liquid behaviour seen in this composition is very robust against pressure and field. By considering the metallurgical aspect of these two samples, we propose that disorder in the U concentration is likely to be responsible for the decrease of T_N with increasing Y concentration, and eventually the appearance of non-Fermi-liquid behaviour in $U_{0.2}Y_{0.8}Pd_3$.

1. Introduction

Current understanding of heavy-fermion compounds is largely based on the Fermi-liquid (FL) description of strongly correlated f electrons [1]. Despite expected strong correlations between f and conduction electrons, one can successfully describe most physical properties of Ce, Yb, and U heavy-fermion compounds in terms of the Abrikosov–Suhl resonance at the Fermi level. However, under certain conditions, some heavy-fermion systems display transport and thermodynamic properties that deviate significantly from what one would expect within the Fermi-liquid description [2–4]. This so-called non-Fermi-liquid (NFL) behaviour has since been observed in many other heavy-fermion systems [5]. As regards the origin of the NFL behaviour, a few theoretical scenarios have been proposed. One is the two-channel Kondo model [2,6], which is thermodynamically equivalent to the quadrupolar Kondo model. Another is a quantum phase transition scenario [3]. According to this theory, when a magnetic transition is suppressed toward $T = 0$ K with pressure, magnetic field, or alloying, the physical properties at this critical point are fundamentally different from the conventional FL predictions. Lastly, there is the Kondo-disorder model which shows that a distribution of Kondo temperatures

can lead to unconventional low-temperature properties [7]. Determining the criterion for the choice of the appropriate model for specific systems has so far proven to be difficult.

$(U_{0.2}Y_{0.8})Pd_3$ was one of the first heavy-fermion compounds shown to exhibit NFL behaviour. The $(U_x Y_{1-x})Pd_3$ pseudobinary alloy system exhibits two crystallographic structures. Above $x = 0.8$, it forms in the dhcp $TiNi_3$-type structure of UPd_3, while for $x \leqslant 0.55$ it adopts the $AuCu_3$-type structure of YPd_3. In between, there is a miscibility gap. With x decreasing from $x = 0.55$, the antiferromagnetic transition temperature is reduced and expected to be at $T = 0$ K for $x = 0.2$. Around this critical concentration, the low-temperature resistivity shows a $-\ln T$ behaviour and the susceptibility a $T^{-\eta}$ one with $\eta = 0.3$. Heat capacity data also show an unusual temperature dependence of $-T \ln T$, and indicate that there should be non-zero entropy at $T = 0$ K. The entropy at $T = 0$ K estimated from the experimental data is $S(T = 0$ K$) = 0.5R \ln 2$. These results were originally interpreted in terms of the two-channel Kondo model [2]. However, this interpretation has been questioned as a result of several experiments including a recent inelastic neutron study [8] suggesting that the ground state of $(U_{0.2}Y_{0.8})Pd_3$ should be a mixture of non-magnetic Γ_3 and magnetic Γ_5, not pure Γ_3 as required in the two-channel Kondo model. Another interpretation of the NFL behaviour of $(U_{0.2}Y_{0.8})Pd_3$ is the $T = 0$ K quantum phase transition scenario [3]. This model seems to be consistent with the fact that the antiferromagnetic transition temperature is driven toward $T = 0$ K as x approaches $x = 0.2$. In relation to the origin of the NFL behaviour of $(U_{0.2}Y_{0.8})Pd_3$, there is an interesting metallurgical study of $(U_x Y_{1-x})Pd_3$ [9]. According to this work, a sample with nominal stoichiometry of $x = 0.2$ shows fluctuations of up to 30% in uranium concentration over a length scale of 10 μm. This problem of inhomogeneity becomes less severe with increasing x. Up to now, it has been largely unknown how this random distribution of U can affect the NFL behaviour.

In the present study, we have investigated the pressure dependence of the Néel temperature of $(U_{0.45}Y_{0.55})Pd_3$ by measuring its resistivity under pressure and magnetic field. For a comparative study, we also examined $(U_{0.2}Y_{0.8})Pd_3$ under the same conditions. Some initial results have been published in [9], but we have subsequently made more extensive measurements to higher pressures using a new steel cell.

2. Experimental details

Two polycrystalline $(U_x Y_{1-x})Pd_3$ samples with $x = 0.2$ and 0.45 were prepared by arc melting high-purity elements: depleted uranium from Ames Laboratory, USA, 5N yttrium, and 4N palladium. To ensure their homogeneity, each sample was flipped and melted 3–5 times. They were then annealed at 800 °C for 24 hours ($x = 0.2$) and one week ($x = 0.45$). Our samples were cut from 10 g melt buttons, which had been used for the neutron scattering experiments [8]. For our discussion later, we note that these samples were examined using electron probe microanalysis (EPMA), which revealed that our $(U_{0.2}Y_{0.8})Pd_3$ had a very large variation in uranium concentration of up to 60% from its nominal stoichiometric value on a length scale of 10–15 μm [11], which is in agreement with a previous report on a similar composition [9]. On the other hand, $(U_{0.45}Y_{0.55})Pd_3$ showed a large grain structure over a length scale of over 100 μm. The variation of uranium concentration is significantly less for $(U_{0.45}Y_{0.55})Pd_3$ than $(U_{0.2}Y_{0.8})Pd_3$. Although annealing improves the inhomogeneity of $(U_{0.2}Y_{0.8})Pd_3$, there is still a large variation in uranium concentration even after heat treatment at 1500 °C [9]. More importantly, the NFL behaviour has been observed mostly in as-cast $(U_{0.2}Y_{0.8})Pd_3$ samples [2, 3], which are likely to be more inhomogeneous than our sample.

Resistivity measurements under pressure were made with Cu–Be and maraging steel pressure cells up to 12 kbar, using a cryostat equipped with a 7 T superconducting magnet. We used a 4:1 mixture of methanol and ethanol as a pressure-transmitting medium. Absolute pressure values were calibrated against the pressure-dependent T_c of Pb. All our measurements were made in the temperature range 2 to 300 K while warming the samples.

3. Experimental data and analysis

In figure 1, we show the resistivity of $(U_{0.45}Y_{0.55})Pd_3$ from 300 to 2 K for five different pressure values. With temperature decreasing from 300 K, the resistivity measured at ambient pressure drops gradually down to 100 K. Below 100 K, it begins to increase and shows a broad hump centred at 34 K. This broad hump corresponds well to the crystal-field splitting of 3 meV between the ground state Γ_3 and the first excited state Γ_5, which was seen in the inelastic neutron scattering experiments [8]. With further cooling, a distinct feature appears at about 20 K, where magnetic susceptibility measurements and elastic neutron diffraction
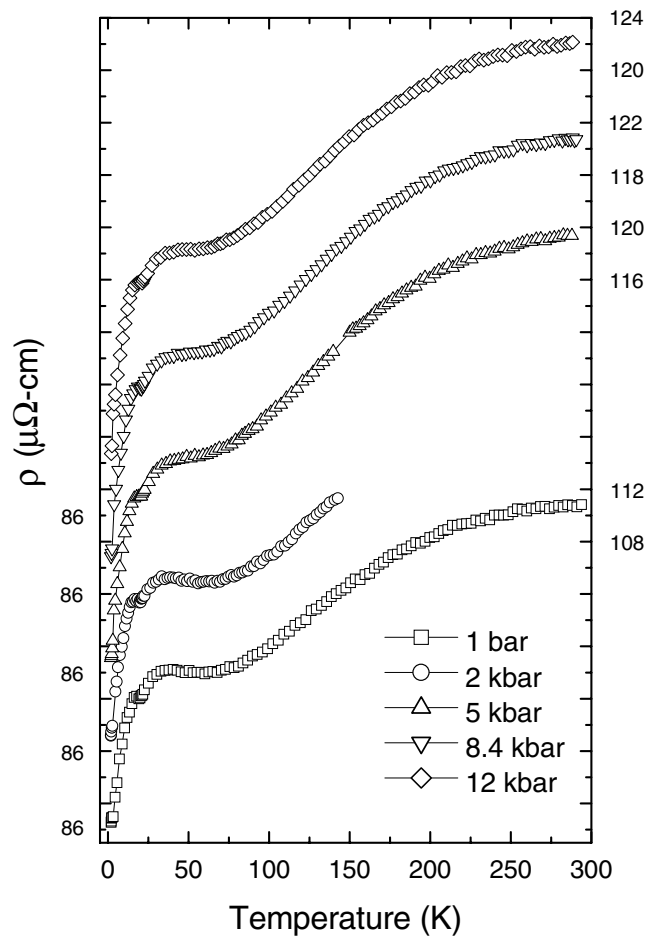

Figure 1. The temperature dependence of the electrical resistivity of $(U_{0.45}Y_{0.55})Pd_3$ from 300 to 2 K for five different pressure values. For clarity, we shifted the data upwards for each pressure.

studies show that long-range antiferromagnetic order develops [11]. With increasing pressure, the resistivity drop between 100 and 300 K becomes bigger. Another noteworthy point in the pressure dependence is that the hump centred at 34 K appears to move slightly toward to higher temperature and become broadened with increasing pressure up to 12 kbar. This increase in the temperature of the hump and the broadening indicate that the low-lying excitation between Γ_3 and Γ_5 actually increases with pressure through hybridization between U 5f electrons and conduction electrons.

As regards the pressure dependence of the antiferromagnetic transition, we show an enlarged picture of the resistivity in figure 2(a). As one can see, the resistivity values increase by 10% at 50 K with pressure from 1 bar to 12 kbar. This increase in the resistivity value with pressure is also in agreement with the earlier conclusion that with increasing pressure, hybridization between U 5f electrons and conduction electrons increases, thus giving rise to higher resistivity. In order to show the pressure dependence of the antiferromagnetic transition more clearly, we show the temperature derivative of the resistivity in figure 2(b). The Néel temperature for the ambient pressure data is marked by the arrow. As can be seen in the figure, the antiferromagnetic transition temperature clearly increases slightly with pressure. The total increase in the Néel temperature between 1 bar and 12 kbar is less than 2 K. The pressure dependence of the Néel temperature is given in figure 3. For comparison, we measured the magnetic field dependence of the Néel temperature and found that there is no shift in T_N up to 7 T within the resolution of our experiment. From magnetoresistance measurements at 4.2 K on $(U_{0.45}Y_{0.55})Pd_3$, we found that the resistivity changes by less than 0.6% from 0 to 17 T.

Regarding the non-Fermi-liquid behaviour seen in $(U_{0.2}Y_{0.8})Pd_3$, a general view is that the Néel temperature is reduced with increasing Y concentration through the shift of the Fermi level. Since U and Y have different valences (U^{4+} and Y^{3+}), increasing Y concentration reduces the Fermi energy thus making the energy difference between the 5f energy level and the Fermi level smaller, i.e. so-called Fermi-level tuning [12]. This tuning can easily explain the decrease in the Néel temperature of (U, Y)Pd$_3$ with increasing Y concentration. However, our data on the pressure dependence of T_N of $(U_{0.45}Y_{0.55})Pd_3$ contradict this explanation: with increasing pressure and hence increasing hybridization between U 5f electrons and conduction electrons, we actually increase, not decrease, the Néel temperature. However, this increase in T_N may be understood if we assume that in $(U_{0.45}Y_{0.55})Pd_3$ the U ions have significantly localized moments as was shown by the inelastic neutron scattering experiments [8]. If the antiferromagnetic transition is due to localized moments coupled through an RKKY-type interaction, then one should expect T_N to increase with enhancing hybridization. Although we acknowledge that the oscillatory nature of the RKKY interaction may complicate this effect, our interpretation of the pressure dependence of T_N does seem to be consistent with the experimental findings. Therefore, we propose that the (U, Y)Pd$_3$ system is located in the weak-interaction regime of the Doniach phase diagram rather than in the strong-interaction regime, as originally thought. This then implies that it is not the variation of hybridization but some other factor that is responsible for the non-Fermi-liquid behaviour of $(U_{0.2}Y_{0.8})Pd_3$. One obvious candidate is the large variation of the U concentration, and in particular close to the critical concentration, as seen by the EPMA study of Y-rich samples. It is a theoretical challenge to calculate exactly how the variation of U concentration affects the transition temperature of (U, Y)Pd$_3$. However, we may reasonably anticipate that the long-range antiferromagnetic order of $(U_{0.45}Y_{0.55})Pd_3$ will be disturbed by locally varying U concentration. As the variations of the U concentration increase, i.e. for sufficiently large fluctuations, the long-range order may not be stable, leading to a metastable state such as the spin-glass state seen in $(U_{0.4}Y_{0.6})Pd_3$ [13]. With a further variation of the U concentration as seen in $(U_{0.2}Y_{0.8})Pd_3$, even the metastable state becomes unsustainable leading to an inhomogeneous magnetic state.
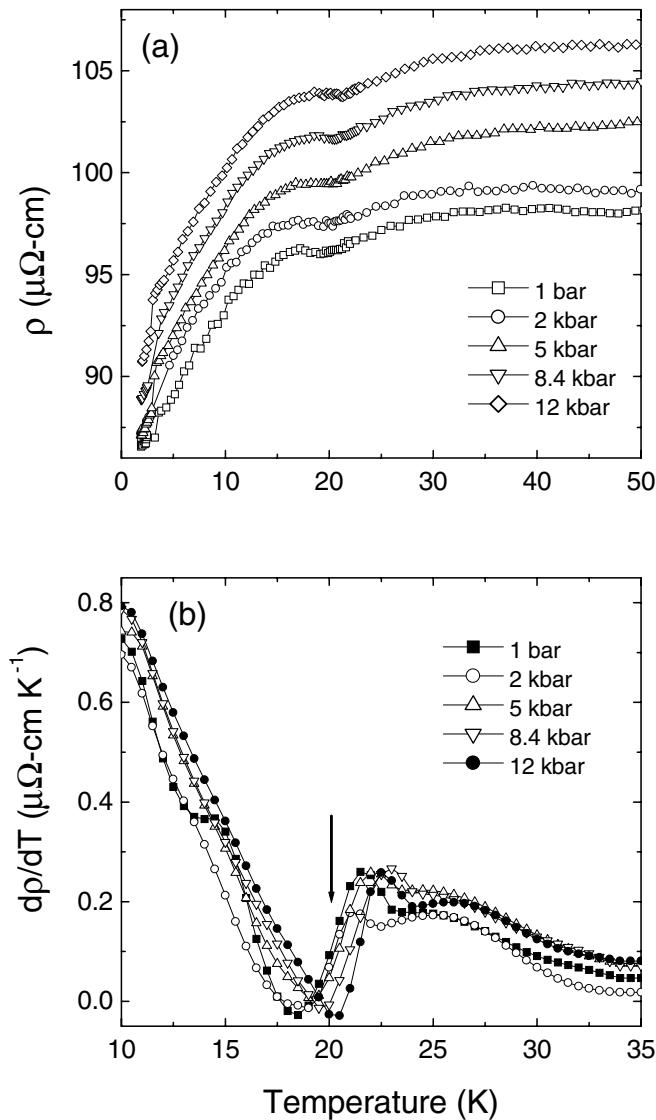
Figure 2. (a) An enlarged picture of the low-temperature resistivity of $(U_{0.45}Y_{0.55})Pd_3$. (b) The temperature derivative of the resistivity showing the pressure dependence of the antiferromagnetic transition temperature. T_N for the ambient pressure data is marked by an arrow.

Considering the $T = 0$ quantum phase transition scenario which explained successfully the non-Fermi-liquid behaviour of $Ce(Cu, X)_6$ with $X = Ag$ and Au [4, 14], we find that this model has some difficulties in being equally successful for the non-Fermi-liquid behaviour seen in $(U_{0.2}Y_{0.8})Pd_3$. Unlike that in $Ce(Cu, X)_6$, the non-Fermi-liquid behaviour seen in $(U_{0.2}Y_{0.8})Pd_3$ is very robust against magnetic field and pressure. Figure 4 shows our resistivity measurements for $(U_{0.2}Y_{0.8})Pd_3$ up to 6.5 kbar and 7 T. As can be seen, there is no noticeable change in the temperature dependence of the resistivity up to 6.5 kbar and 7 T: the pressure and field dependence of the non-Fermi-liquid behaviour of $(U_{0.2}Y_{0.8})Pd_3$ is totally different from
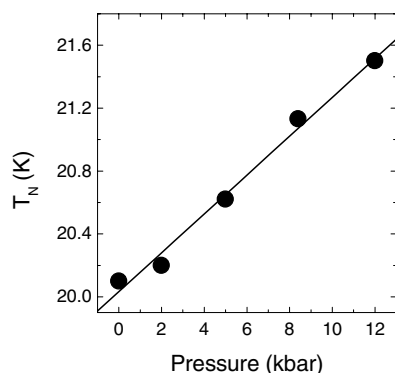
Figure 3. The pressure dependence of the Néel temperature of $U_{0.45}Y_{0.55}Pd_3$ up to 12 kbar. The line is a guide to the eye.
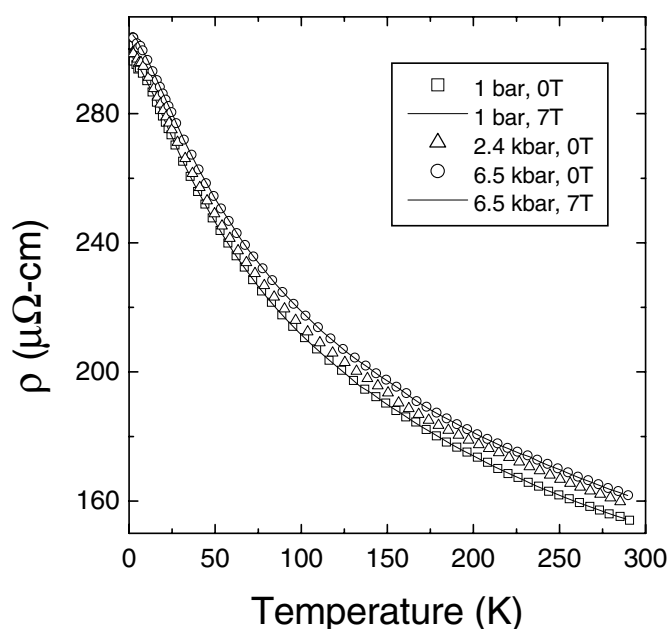

Figure 4. The temperature dependence of the electrical resistivity of $(U_{0.2}Y_{0.8})Pd_3$ measured up to 6.5 kbar and 7 T.

that of the $Ce(Cu, X)_6$ systems. This difference clearly demonstrates the different origins of the non-Fermi-liquid behaviour of $(U_{0.2}Y_{0.8})Pd_3$ and that of $Ce(Cu, X)_6$.

Any theory concerning the origin of the non-Fermi-liquid behaviour of $(U_{0.2}Y_{0.8})Pd_3$ has to be able to explain both its field and pressure dependence as well as its possible connection with the metallurgical problem concerning the variation of U concentration. It should also take into account our finding that the Néel temperature of $(U_{0.45}Y_{0.55})Pd_3$, where there is very little variation in U concentration, increases rather than decreases with pressure. Among currently available theories, a newly proposed theoretical model invoking the Griffiths phase [15] seems to be most compatible with all these experimental results. In this theory, studies of the effects of disorder on magnetic phase transitions show that the disordered-Kondo-lattice model can

be mapped onto the random Ising model in a random transverse magnetic field. With certain disorder, this model predicts an inhomogeneous magnetic phase at $T = 0$ K, which causes the non-Fermi-liquid behaviour at low temperature. In fact, this theoretical model has been found to fit the low-temperature data for the heat capacity and magnetization of $U_{0.2}Y_{0.8}Pd_3$ better than the original two-channel Kondo model [16]. As we have demonstrated in this paper, our experimental data for $(U, Y)Pd_3$ point toward the importance of disorder. Since the disorder present in $(U, Y)Pd_3$ is of metallurgical origin, we expect that there would be a very small dependence of the non-Fermi-liquid behaviour on pressure or field, as we have indeed found.

In conclusion, to understand the non-Fermi-liquid behaviour of $(U_{0.2}Y_{0.8})Pd_3$, and in particular its relation with a large variation of U concentration seen in Y-rich samples, we have investigated the pressure and field dependence of the resistivity of $(U_x Y_{1-x})Pd_3$ with $x = 0.2$ and 0.45. By studying $(U_{0.45}Y_{0.55})Pd_3$, which was shown to be more homogeneous from our EPMA study than $(U_{0.2}Y_{0.8})Pd_3$, we have found that the Néel temperature of $(U_{0.45}Y_{0.55})Pd_3$ actually increases with increasing hybridization between the 5f electrons and conduction electrons. We also discussed why the non-Fermi-liquid behaviour of $(U_{0.2}Y_{0.8})Pd_3$ is very robust against pressure and field. By considering these results, along with the metallurgical problem of U concentration, we propose that the non-Fermi-liquid behaviour of $(U_{0.2}Y_{0.8})Pd_3$ can be understood in terms of the inhomogeneous magnetic phase at $T = 0$ K. We hope that our results will stimulate further theoretical studies of the relationship between concentration fluctuations and NFL phenomena.

Acknowledgments

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