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ELECTRICAL RESISTIVITY OF YbInAu₂ AND YbCuAl UP TO 8 GPa

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The electrical resistivity $\rho(T)$ of the intermediate valence compounds YbInAu₂ and YbCuAl was investigated up to 8 GPa. With increasing pressure, the Kondo temperature decreases and the residual resistivity ρ_0 increases. At low temperature, $\rho(T)$ shows a Fermi liquid behaviour in YbCuAl whereas it develops a minimum in YbInAu₂. These findings are discussed in terms of the Kondo lattice disorder. © 1998 Elsevier Science Ltd. All rights reserved

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1. INTRODUCTION

Intermetallic compounds with Ce or Yb rare earth components can exist in the intermediate valence (IV) state. Such metals show a variety of anomalies in their physical properties with a characteristic temperature. Above this temperature, a local description of the $4f$ -electron seems applicable, whereas an itinerant description appears more adequate at very low temperature. In the Yb-based IV compounds, the Yb-valence takes values between +2 and +3, corresponding to the non-magnetic $4f^{14}$ and the magnetic $4f^{13}$ configurations of the free Yb-ion, respectively. The volume of the latter configuration is smaller than that of the former [1]. Thus, the application of pressure, i.e. the volume reduction, on the Yb-compounds favours a valence state +3 and hence the magnetically ordered state. This pressure effect was well confirmed in the case of the IV compound YbCu₂Si₂ which transits to the magnetic state at about 8 GPa [2]. In the case of the other Yb-compounds studied by the electrical resistivity, no clear signs of magnetic transition are found [3, 4].

At ambient pressure, the Yb-valence in YbInAu₂ (cubic CsCl-type) is 2.68 [5] whereas it is 2.96 [6] in YbCuAl (hexagonal Fe₂P-type). The latter compound can be considered to be closer to the magnetic state

than the former. The coefficient γ is about 40 [5] and 267 mJ/mol K² [7], for YbInAu₂ and YbCuAl, respectively. Here we report electrical resistivity $\rho(T)$ investigations for YbCuAl and YbInAu₂ in between 1.2 K < T < 300 K and up to a pressure of 8 GPa. At the highest pressure, the resistivity was measured down to 30 mK and in a magnetic field ($B \leq 8$ T).

2. EXPERIMENT AND RESULTS

Polycrystalline samples were prepared by melting stoichiometric amounts in a sealed tantalum crucible [8] and by post-annealing at 800°C for several days. Measurements of the electrical resistivity were performed with the four point method. A Bridgman-type high pressure cell with steatite as pressure transmitting was used [8]. The two compounds were measured simultaneously in one pressure cell. The pressure was measured by a Pb-wire [9], playing also the role of electrical contact between the two samples [8].

Figure 1 shows the temperature dependence of the electrical resistivity $\rho(T)$ of YbInAu₂ at selected pressures up to 8 GPa. The value of ρ at 300 K increases from 47 $\mu\Omega$ cm at $P = 0$ to 53 $\mu\Omega$ cm at $P = 4.7$ GPa and then decreases down to 51 $\mu\Omega$ cm at 8 GPa. This pressure variation coincides with the development of a maximum in $\rho(T)$ as is evident at 30 K for $P = 8$ GPa. The value of the resistivity at 1.2 K increases from 9 $\mu\Omega$ cm at $P = 0$ GPa to 44 $\mu\Omega$ cm at 8 GPa, i.e. an increase by a factor 5, indicating an increase of the

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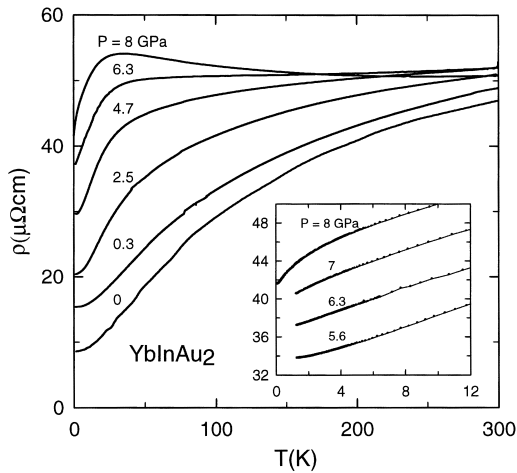


Fig. 1. Electrical resistivity $\rho(T)$ of YbInAu_2 at selected pressures. The inset shows the change of the curvature at low T .

residual resistivity ρ_0 . At low temperature, the curves show a change of curvature between 5.6 and 7 GPa (see inset Fig. 1). At $P = 6.3$ GPa, the curve presents a linear behaviour below 10 K. The detailed analysis of the curves at low temperature revealed the development of a minimum at low temperature for $P > 2.5$ GPa as is shown in Fig. 2 for $P = 3.6$ GPa and in the inset for $P = 8$ GPa. The position of this minimum shifts to lower temperature upon pressure increase (2.2 K at 3.6 GPa and 0.1 K at 8 GPa). Such a minimum also develops with pressure in the case of CeCu_2Si_2 [10]. No sign of magnetic order is detected at low temperature and at high pressure. The magnetoresistivity measured at 8 GPa is negative,

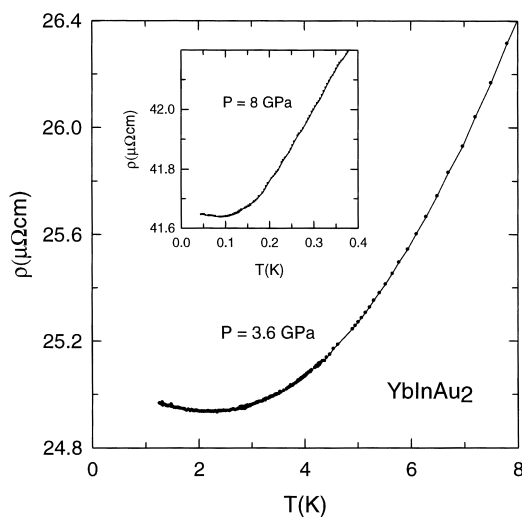


Fig. 2. The appearance of the minimum in the low T $\rho(T)$ -curves above 2.5 GPa. The position of the minimum shifts to lower temperature at 8 GPa (inset).

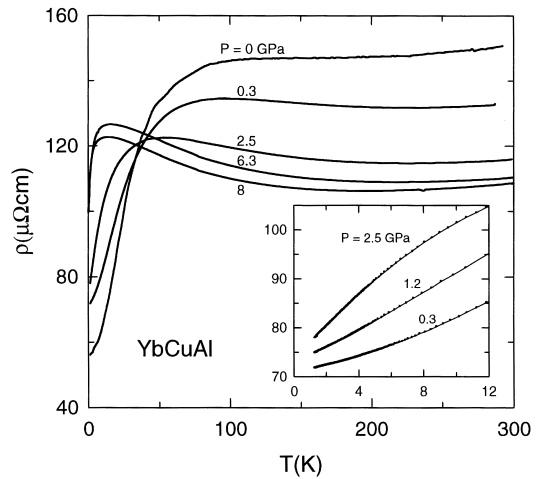


Fig. 3. Electrical resistivity $\rho(T)$ of YbCuAl at representatives pressures. The inset shows the change of the low T behaviour.

i.e. the resistivity decreases by $\sim 10\%$ at 8 T in the temperature range $30 \text{ mK} < T < 4.2 \text{ K}$.

The electrical resistivity $\rho(T)$ of YbCuAl for pressures up to 8 GPa is displayed in Fig. 3. The curves show a similar behaviour as that reported in [11]. The resistivity at 300 K shifts down on increasing pressure ($152 \mu\Omega \text{ cm}$ and $108 \mu\Omega \text{ cm}$ at $P = 0$ and 8 GPa, respectively). This behaviour is in contrast to the pressure dependence of the $\rho(T)$ -value at 1.2 K which increases from $57 \mu\Omega \text{ cm}$ at $P = 0$ to about $100 \mu\Omega \text{ cm}^{-1}$ at 8 GPa. A maximum, attributed to the Kondo effect, develops in $\rho(T)$ with pressure and is clearly seen at 6.3 GPa (see Fig. 3). The position of this maximum decreases and is nearly unchanged at higher pressures. Below this maximum, the resistivity curves present a change of curvature in the P -range $0.3 \text{ GPa} < P < 2.5 \text{ GPa}$ as illustrated in the inset of Fig. 3. Like in YbInAu_2 , the resistivity of YbCuAl at 1.2 GPa presents a linear T -dependence below 10 K. Below 1.2 GPa, the curves can be fitted with $\rho(T) = \rho_0 + AT^2$ for $T < T_A$, with A and T_A depending on pressure. Because of the fact that the limit temperature T_A shifts rapidly to lower temperature ($T_A < 1.2 \text{ K}$), it was not possible to see this behaviour for $P > 1.2 \text{ GPa}$. A quadratic temperature dependence of $\rho(T)$ is seen again at 8 GPa which is depicted for T down to 30 mK in Fig. 4. An increase of A from 0.087 to $8.01 \mu\Omega \text{ cm K}^{-2}$ between 0 and 8 GPa is found, i.e. an increase by a factor 92. At 8 GPa, the curve presents a clear change of slope at 1 K which could be interpreted as a sign of a magnetic transition. At this pressure, the magnetoresistivity is also negative below 4.2 K ($\rho(T)$ is reduced by $\sim 12\%$ in a magnetic field of 8 T). The anomaly at 1 K seems to disappear at 3 T, indicating the possibility of antiferromagnetic ordering (inset of Fig. 4).

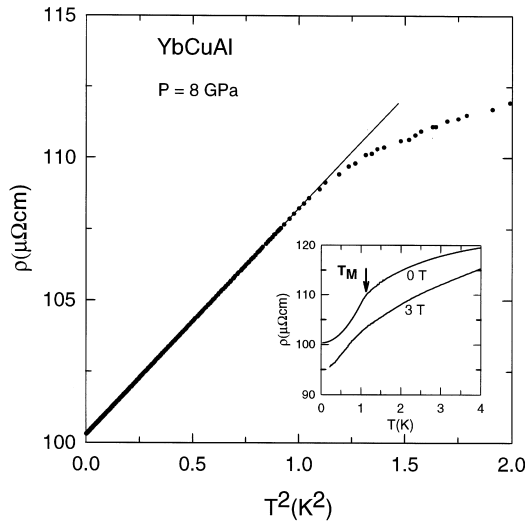


Fig. 4. The T^2 -dependence of $\rho(T)$ in YbCuAl for $P = 8$ GPa. A clear change of slope is observed at 1 K. The inset shows the curve at $B = 0$ and 3 T. The anomaly at 1 K is interpreted as a sign of a magnetic order and seems to disappear in a magnetic field of 3 T.

3. DISCUSSION

At high temperature the total resistivity $\rho(T)$ in the presence of the Kondo effect can be written as:

$$\rho(T) = \rho_0 + \rho_{\text{mag}}(T) + \rho_{\text{ph}}(T) = \rho_1 - c \ln T + aT \quad (1)$$

where ρ_0 , $\rho_{\text{mag}}(T)$ and $\rho_{\text{ph}}(T)$ describe the scattering of imperfections, spins and phonons, respectively. In this equation, a and c are constants and ρ_1 is the sum of ρ_0 and the spin disorder resistivity associated to the Kondo exchange interaction [12]. Fitting the right hand side of equation (1) to the $\rho(T)$ -data at high temperature, a pressure-independent phonon contribution a is found. Subtracting this contribution from $\rho(T)$ gives the residual and magnetic part $\rho_0 + \rho_{\text{mag}}(T)$. The curves obtained show a maximum at T_{max} , which is plotted as a function of pressure in Fig. 5. For the two compounds T_{max} decreases strongly with pressure. An interpretation for this strong decrease can be given in the picture where T_{max} is of the order of the Kondo temperature $T_K \propto \exp(-1/J \cdot n(E_F))$, with J the exchange coupling constant and $n(E_F)$ the density of states of the conduction electrons at the Fermi level. Assuming that $n(E_F)$ is not affected or only weakly affected by pressure, the enormous decrease of T_{max} (or T_K) can be interpreted by a reduction of the exchange coupling J . The logarithmic slope c , indicating incoherent Kondo scattering processes on the excited crystal field (CF) levels, does not change with pressure as for other Yb compounds, e.g. YbCu₂Si₂ [2], YbCu_{4.5} [3], in contrast to many Ce compounds [13]. The slope c is determined by contributions due to $n(E_F)$, the s - f

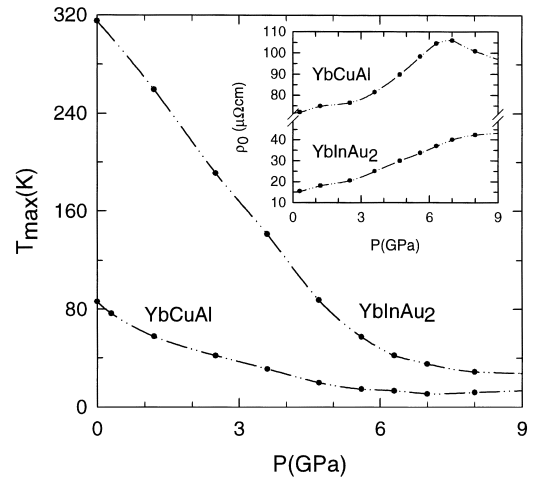


Fig. 5. The temperature T_{max} , where the magnetic resistivity presents a maximum and the residual resistivity ρ_0 (inset) as a function of pressure for YbInAu₂ and YbCuAl. The two quantities have an opposite pressure dependence.

coupling constant J and the CF effect [12]. The application of pressure on the Yb-compounds is similar to a decrease of the product $J \cdot n(E_F)$. To get a pressure-independent logarithmic slope c , CF effects have to be considered. The CF splitting could undergo a reduction with increasing pressure because of a redistribution of conduction electrons [14].

In the vicinity of the critical point, i.e. between the magnetic and the non-magnetic phase in Doniach's diagram [15], the existence of a non-Fermi liquid (NFL) behaviour, characterised by a $\rho = \rho_0 + AT^\alpha$ ($\alpha < 2$) dependence of the low temperature electrical resistivity, is discussed. On approaching this region, a cross-over from a FL ($\alpha = 2$) to a NFL behaviour is predicted and the A coefficient diverges [16]. Furthermore, at the critical point, the NFL behaviour should be seen for $T < T_1$ and followed by a linear $\rho(T)$ dependence for $T_1 < T < T_2$ [16]. This could be an interpretation of the linear $\rho(T)$ behaviour at 6.3 and 1.2 GPa for YbInAu₂ and YbCuAl, respectively (insets of Fig. 1 and Fig. 3). However, in the case of YbCuAl, the $\rho(T)$ curves at very low temperature confirm rather a FL description (Fig. 4). Therefore, these $\rho(T)$ -dependencies can not be considered as clear indications of a NFL behaviour, as reported in the case of CeNi₂Ge₂ [17].

The pressure dependence of the residual resistivity ρ_0 obtained by extrapolating the resistivity to $T = 0$ K is reported in the inset of Fig. 5. For the two compounds, ρ_0 increases rapidly on increasing pressure. A maximum in $\rho_0(P)$ of YbCuAl is found around 7 GPa and probably above 8 GPa for YbInAu₂. This maximum can be correlated to a magnetic ordering in comparison to YbCu₂Si₂ which shows a pronounced maximum in

$\rho_0(P)$ at the magnetic instability [2]. To describe this ρ_0 behaviour and the pressure induced minimum of the low temperature for YbInAu₂, we proposed [2] to approximate the resistivity at low temperature ($T < T_K$) by

$$\rho(P, T) = \rho_{0L} + A(P)T^2 + \rho_H(P, T), \quad (2)$$

with an additional contribution ρ_H describing the disorder in the Kondo lattice. The static contribution ρ_{0L} is pressure and temperature independent and reaches values of typically 1–10 $\mu\Omega$ cm for high quality crystals. The positive contribution $A(P)T^2$ describes the coherent scattering of the quasi-particles of the perfect Kondo lattice [18]. The coefficient A is related to T_K by $T_K \propto \gamma^{-1} \propto A^{-1/2}$. The third term can be written as $\rho_H(P, T) = \rho_{0H} - BT^m$, with a temperature independent ρ_{0H} and a coefficient $B > 0$. If m is close to 2, the observed quadratic coefficient ($A - B$) is clearly less than the value deduced from the empirical ratio $A/\gamma^2 = 10^{-5} \mu\Omega \text{ cm K}^2 \text{ mol}^2/\text{mJ}^2$ [19] which was found for several Ce and U compounds. In particular, for YbCu₄Ag [4] and YbCu_{4.5} [3] A/γ^2 is in the range $0.02 - 0.03 \times 10^{-5} \mu\Omega \text{ cm K}^2 \text{ mol}^2/\text{mJ}^2$. Furthermore, a minimum appears in the total resistivity if the term $-BT^m$ dominates, like in YbInAu₂ and CeCu₂Si₂ [10]. On the other hand, ρ_0 and hence ρ_{0H} , increase with pressure as it was shown for YbCu₂Si₂ [2]. The correlation with the magnetic instability is the reason to attribute the term ρ_{0H} to an effect due to Kondo holes. These holes scatter like occupied Kondo sites [20]. A disorder effect like this might screen phenomena in the low temperature resistivity, as a magnetic order, e.g. in YbInAu₂, YbCu₄Ag [4] and YbCu_{4.5} [3], a NFL behaviour, e.g. in YbCuAl and YbCu₂Si₂ [2], or even superconductivity, e.g. in CeCu₂Si₂ [10] and CePd₂Si₂ [21, 22].

4. CONCLUSION

The effect of pressure on the electrical resistivity $\rho(T)$ of the intermediate valence compounds YbInAu₂ and YbCuAl was investigated up to 8 GPa. With increasing pressure, the temperature T_{\max} , where $\rho_{\max}(T)$ shows a maximum, decreases strongly, indicating the decrease of the Kondo temperature. At low temperature, a Fermi liquid behaviour ($\rho = \rho_0 + AT^2$) is found in YbCuAl, with a strongly increasing $A(P)$ coefficient. In the case of YbInAu₂, $\rho(T)$ develops a minimum at low temperature, whose position shifts down with pressure. The pressure dependence of the low temperature resistivity was interpreted in the context of the Kondo hole effect which leads to an increase of the residual resistivity ρ_0 on approaching the magnetic instability. The maximum

in $\rho_0(P)$ around 7 GPa and the $\rho(T)$ anomaly at 1 K (8 GPa) in YbCuAl are interpreted as signs of magnetic order.

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