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Low-temperature resistivity of $\text{YbCu}_{5-x}\text{Au}_x$ under magnetic fields

N Tsujii¹, T Terashima¹, C Terakura¹, G Kido¹, T Kawabata²,
K Yoshimura² and K Kosuge²

¹ National Research Institute for Metals, 1-2-1 Sengen, Tsukuba, Ibaraki 305-0047, Japan

² Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan

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Abstract

YbCu_5 shows a typical heavy-fermion behaviour while YbCu_4Au exhibits an antiferromagnetic transition below 1 K. We have investigated the low-temperature electrical resistivity, ρ , of the pseudo-binary compound $\text{YbCu}_{5-x}\text{Au}_x$ ($0 \leq x \leq 1.0$) as functions of temperature (T) and magnetic field (H). The $\rho(T)$ data indicate that the magnetic transition occurs for $x = 1.0, 0.8$, and 0.6 at $T_{\text{order}} = 0.6, 0.4, 0.1$ K, respectively, whereas magnetic ordering is absent for $x \leq 0.2$ down to the lowest temperature. $\rho(H)$ for $x = 0, 0.1$, and 0.2 measured at 0.05 K shows a characteristic behaviour in the non-magnetic Kondo-lattice state. It has been found that the increase of the Au concentration x results in a rapid decrease of the Kondo temperature, T_K , as well as the evolution of impurity Kondo behaviour, destroying the Kondo-lattice state for $x = 0$. This may imply that the Au substitution causes a significant change of the local electronic structures. $\rho(T)$ for $x = 0.6$ varies nearly as $T^{3/2}$ at 0.5 T below 0.15 K, while it shows a T^2 -power law at 1 T. This behaviour can be interpreted as the crossover caused by a magnetic field from a magnetic ordered state at zero field through a non-Fermi-liquid state to the Fermi-liquid state at high field.

1. Introduction

The YbCu_4M ($\text{M} = \text{In}, \text{Ag}, \text{Au}$, etc) compounds with the cubic AuBe_5 -type structure have been widely studied because of their variety of physical properties [1, 2]. A first-order valence transition occurs in YbCu_4In at the temperature $T_V = 40$ K [3] or at the magnetic field $H_V = 35$ T [4]. In YbCu_4Ag , a typical Kondo-lattice state is realized with a non-magnetic Fermi-liquid ground state [5, 6]. The thermodynamic properties are well described by the Bethe-ansatz solution of the Coqblin-Schrieffer model [7] with a single energy parameter $T_0 = 160$ K [5, 8], where T_0 is proportional to the Kondo temperature, T_K . Neutron scattering experiments revealed a broad quasi-elastic linewidth and found no evidence of a well-defined

crystal-field excitation for YbCu₄Ag [9]. On the other hand, the value of T_K for YbCu₄Au is so small that this system undergoes a magnetic ordering below 1 K [5]. Moreover, neutron scattering experiments revealed the presence of clear crystal-field excitations as well as a very narrow quasi-elastic linewidth for YbCu₄Au [9], in a sharp contrast to the case for YbCu₄Ag. The magnetic structure below 1 K is suggested to be a very complicated one by the findings of a neutron diffraction experiment [10].

In addition to these YbCu₄M systems, high-pressure synthesis has revealed the existence of cubic AuBe₅-type YbCu₅ [11, 12], instead of the hexagonal YbCu₅ phase which was reported to exist at ambient pressure [13]. Cubic YbCu₅ (hereafter simply referred to as ‘YbCu₅’) is a typical heavy-fermion system with a large electronic specific heat coefficient, $\gamma = 550 \text{ mJ mol}^{-1} \text{ K}^{-2}$ [12]. The characteristic temperature of YbCu₅ was estimated as $T_0 = 60 \text{ K}$ from magnetic susceptibility and specific heat data [12]. Since YbCu₅ can be regarded as the mother material for all the cubic YbCu₄M compounds, investigating the pseudo-binary systems, YbCu_{5-x}M_x ($0 \leq x \leq 1.0$), is useful as regards understanding the mechanism of evolution of the various physical properties of YbCu₄M. Detailed experiments have been performed for YbCu_{5-x}M_x with M = In and Ag [14–17]. For YbCu_{5-x}Ag_x in particular, Fermi-liquid behaviour is observed over the whole range of Ag concentration x [15, 17], and the variation with Ag concentration of T_0 is found to be well explained by the chemical pressure effect [12, 18], in which external pressures suppress T_0 for Yb-based compounds. In other words, the high characteristic temperature of YbCu₄Ag is mainly attributable to the lattice volume of YbCu₄Ag being larger than that of YbCu₅.

On the other hand, T_K for YbCu₄Au is much smaller than that of YbCu₅ in spite of the former having a larger lattice constant than the latter. This fact indicates that the chemical pressure effect does not play an important role for the Au-substituted compounds. One possible way of explaining this is to take the crystal-field effect into consideration. Neutron scattering [9] and electrical resistivity measurements under pressure [19] have revealed the existence of a clear crystal-field splitting in YbCu₄Au. Its effect must contribute to the reduction of T_K for YbCu₄Au. On the other hand, it has been suggested, on the basis of a self-consistent calculation of the electronic structure of these compounds, that the f-d hybridization strength differs substantially between YbCu₄Ag and YbCu₄Au, resulting in the different ground states [20]. Therefore, it is of considerable interest to examine how T_K changes with the change of the Au concentration in the pseudo-binary YbCu_{5-x}Au_x.

Moreover, the critical region between the magnetically ordered and the paramagnetic Kondo state has been attracting much interest. Many anomalous phenomena such as non-Fermi-liquid (NFL) and non-BCS superconductivity have been observed in the vicinity of the critical point. Recently, NFL behaviour has been reported in several Yb-based systems, such as YbCu_{3.5}Al_{1.5} [21] and YbRh₂Si₂ [22]. In this respect, YbCu_{5-x}Au_x is also interesting, since we can reach the critical point for a finite value of x .

The aim of this paper is to clarify the ground state of YbCu_{5-x}Au_x and to discuss the origin of the magnetic ordering in YbCu₄Au. We have performed low-temperature measurements of the electrical resistivity (ρ) for YbCu_{5-x}Au_x ($0 \leq x \leq 1.0$) as functions of temperature (T) and magnetic field (H). Our results indicate that the value of T_K decreases systematically with increasing x , resulting in a magnetically ordered state for $x \geq 0.6$. We can expect the quantum critical point to lie around the Au concentration $x = 0.4$ – 0.6 . The physical properties near the quantum critical state are considered to be dominated by very low-energy magnetic excitations, and are quite sensitive to external parameters such as magnetic field and pressure. We have hence performed $\rho(T)$ measurements for $x = 0.4$ and 0.6 under magnetic fields. We will give the experimental results in section 3, and then summarize the electrical properties of the YbCu_{5-x}Au_x system in section 4.

2. Experimental procedure

Polycrystalline samples of $\text{YbCu}_{5-x}\text{Au}_x$ with $x = 1.0$ (YbCu_4Au), 0.8, 0.6, 0.4, 0.2 were prepared by using an argon-arc furnace and subsequent annealing. The samples with $x = 0.1$ and 0 (YbCu_5) were synthesized by the high-pressure technique under 1.5 GPa [12]. The electrical resistivity was measured by an ac four-probe method. The error in the absolute value of the resistivity is less than $\pm 20\%$, and mainly arises from the estimation of the sample dimensions. The temperature dependence of the resistivity for $0.1 \leq x \leq 1.0$ was measured from 0.03 K to 1.5 K using a ^3He - ^4He dilution refrigerator, while the resistivity for $x = 0$ was measured above 0.2 K. The magnetic field of up to 18 T was generated by a superconducting magnet. The direction of the electric current was set parallel to the field.

3. Results and discussion

3.1. The temperature dependence of the resistivity of $\text{YbCu}_{5-x}\text{Au}_x$

In figure 1, the temperature dependence of the electrical resistivity, $\rho(T)$, of $\text{YbCu}_{5-x}\text{Au}_x$ is shown on a logarithmic temperature scale. The data shown are normalized by the values at 1 K. $\rho(T)$ for $x = 1.0$ (YbCu_4Au) decreases rapidly below $T = 1.3$ K, due to the onset of antiferromagnetic ordering, as has been verified by several authors [5, 10]. The Néel temperature can be estimated to be the temperature at which $d\rho/dT$ has a maximum, which yields the value $T_N = 0.7$ K for YbCu_4Au . This is close to the value that has been obtained from the specific heat data [5].

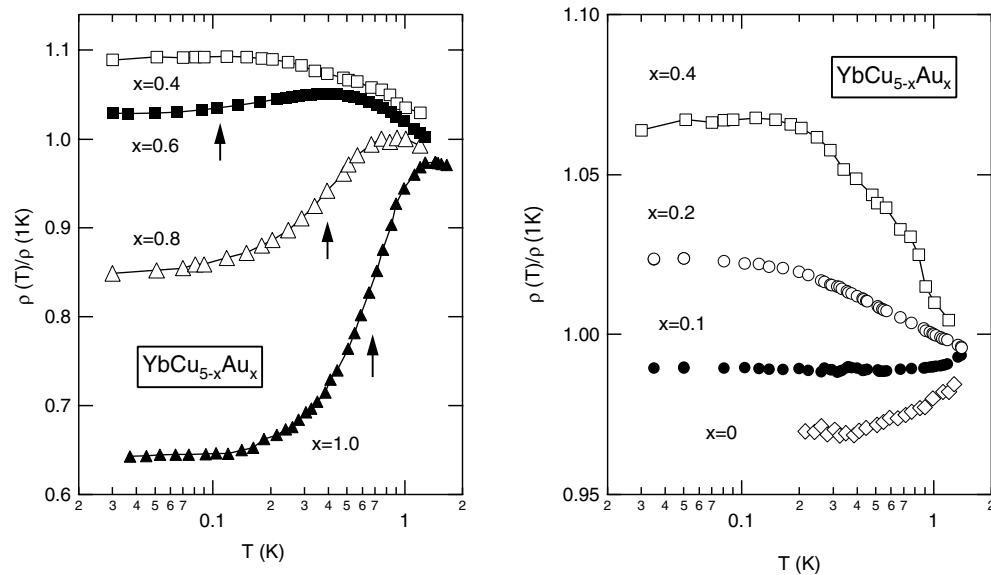


Figure 1. The temperature dependence of the electrical resistivity of $\text{YbCu}_{5-x}\text{Au}_x$. The resistivity shown is normalized by the value at 1 K. The magnetic ordering temperature, T_{order} , is indicated by an arrow. T_{order} has been estimated as the temperature at which $d\rho/dT$ has a maximum.

For $x = 0.8$ and 0.6, the $\rho(T)$ curves show maxima at $T_{max} = 0.80$ K and 0.42 K, respectively. These maxima imply the onset of magnetic ordering in these compounds. We have evaluated the magnetic ordering temperature, T_{order} , as the temperature at which $d\rho/dT$ has a

maximum, and have obtained $T_{order} = 0.40$ K and 0.10 K for $x = 0.8$ and 0.6, respectively. For $x = 0.4$, one can find a very faint maximum in figure 1. It is not clear whether this maximum is due to magnetic order or not, since this maximum is very broad and the $d\rho/dT$ curve does not show a maximum down to 0.03 K. To establish whether magnetic order is present, other experiments, such as specific heat and magnetic susceptibility investigations, should be performed.

For $x \leq 0.2$, no anomalies are observed in the $\rho(T)$ data down to the lowest temperatures. $\rho(T)$ for $x = 0$ becomes almost constant below 0.5 K. It has been demonstrated that YbCu₅ ($x = 0$) has a non-magnetic Kondo-lattice state above 1.6 K on the basis of specific heat [12] and NQR measurements [23]. Considering the data in figure 1, it is concluded that YbCu₅ has a non-magnetic Kondo-lattice state down to 0.2 K.

$\rho(T)$ for $x = 0.2$ increases gradually as temperature decreases. As one can see in figure 1, the increase of $\rho(T)$ is proportional to $-\ln T$ below 1.4 K down to about 0.2 K, until it reaches a constant value below 0.1 K. This behaviour resembles the incoherent Kondo effect due to magnetic impurities. So one may conclude that an impurity Kondo state occurs in the $x = 0.2$ system, not a coherent Kondo-lattice one. In this case, the ground state is the local Fermi-liquid state, where individual magnetic ions are coupled with carrier electrons and form a spin-singlet state. However, we should mention that the electrical resistivity for $x = 0.2$ shows a T^2 -dependent decrease above 2 K [24], a characteristic behaviour of a Kondo-lattice system. Therefore, both Kondo-lattice formation and the impurity Kondo effect should be taken into consideration to understand the electronic state for $x = 0.2$. It should be noted that the impurity Kondo-like behaviour is also observed in the YbCu₄Ag_{1-y}Au_y system at around $y = 0.3$ [25]. In this pseudo-binary system, the Kondo-lattice state of YbCu₄Ag is quickly destroyed by the substitution of Au. These facts indicate that Au substitution destabilizes the Kondo-lattice state, possibly due to a large change of the local electronic structure.

For the $x = 0.1$ system, $\rho(T)$ is almost temperature independent below 1 K. Since $\rho(T)$ for $x = 0.1$ also shows a T^2 -behaviour above 2 K [24], this system is classified as a Kondo-lattice system with a non-magnetic state down to 0.03 K.

3.2. The field dependence of the resistivity of YbCu_{5-x}Au_x

In figure 2, we show the field dependence of the resistivity of YbCu_{5-x}Au_x. The data shown are normalized by the value at $H = 0$ T. $\rho(H)$ for $x = 0$ (YbCu₅) increases monotonically with fields up to 17 T. $\rho(H)$ for $x = 0.1$ and 0.2 shows maxima around $H_m = 8$ T and 4 T, respectively. Positive magnetoresistance or a maximum in $\rho(H)$ is often observed in non-magnetic Kondo-lattice systems. For instance, $\rho(H)$ for YbNi₂Si₂ increases monotonically with field [26]. YbCu₄Ag [27] and YbNi₂B₂C [28] show maxima in their $\rho(H)$ curves at $H_m = 32$ T and 8 T, respectively. Theoretical calculation based on the periodic Anderson model also predicts the $\rho(H)$ curve of a Kondo-lattice system to have a maximum at $H = H_m$, whose value is proportional to that of the Kondo temperature, T_K [29]. Thus $\rho(H)$ for $x = 0, 0.1$, and 0.2 in figure 2 can be regarded as characteristic behaviour for Kondo-lattice systems with non-magnetic ground states. This interpretation is consistent with the T^2 -dependence of $\rho(T)$ for these systems observed above 2 K [24]. We can see in figure 2 that the value of H_m decreases with increasing x . The value of H_m for $x = 0$ is larger than 17 T, while those for $x = 0.1$ and 0.2 are 8 T and 4 T, respectively. This suggests that the value of T_K decreases rapidly with increase of x . In contrast to the case for YbCu_{5-y}Ag_y, the decrease of T_K for YbCu_{5-x}Au_x cannot be explained by the chemical pressure, since the lattice volume increases with increasing x [16]. For the $\rho(H)$ curve for $x = 0.2$ in figure 2, one also notices a minimum at around $H = 1.5$ T, which we discuss later.

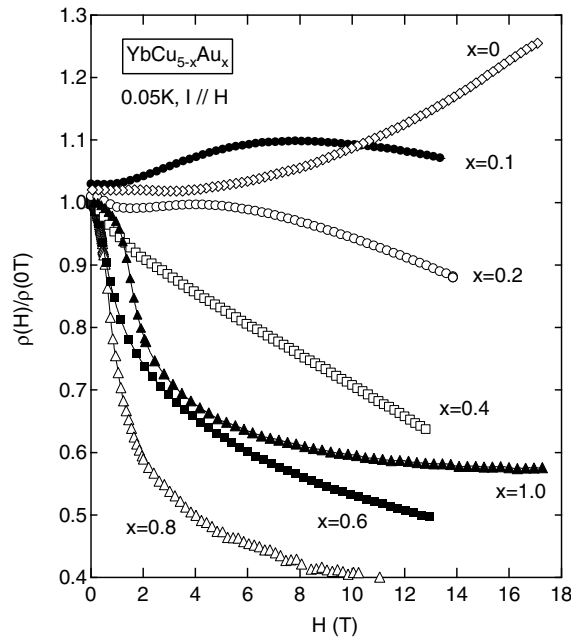


Figure 2. The field dependence of the resistivity of $\text{YbCu}_{5-x}\text{Au}_x$ measured at 0.05 K. The resistivity shown is normalized by the value at $H = 0$ T. The data are shifted vertically for clarity.

For systems with $x \geq 0.4$, $\rho(H)$ decreases with increasing field. A large negative magnetoresistance is often observed in antiferromagnetic Kondo systems around T_N [30]. In our case, $\rho(H)$ decreases most rapidly for the $x = 0.8$ system. The magnetoresistance ratio $[\rho(H) - \rho(0)]/\rho(0)$ for $x = 0.8$ reaches -50% at 4 T. This large magnetoresistance implies a strong coupling between the 4f moment and the conduction electrons. One finds that $\rho(H)$ for $x = 0.4$ decreases linearly over a wide range of magnetic field.

In figure 3, the $\rho(H)$ curves for $x = 1.0$ measured at 0.05 K and 1.8 K are shown. $\rho(H)$ at 1.8 K decreases quite rapidly with field, and the magnetoresistance ratio is about -50% at $H = 5$ T. At a high magnetic field of $H > 10$ T, the $\rho(H)$ values at 0.05 K and 1.9 K are almost the same: $\simeq 9 \mu\Omega \text{ cm}$. Hence, this value can be regarded as the contribution of the lattice defects and extrinsic impurities, without a magnetic moment.

In figure 4(a), we show the low-field region of $\rho(H)$ for $x = 1.0$ measured at various temperatures. The $\rho(H)$ curves are almost flat below about 1 T, and decrease steeply above 1 T. Similar behaviour is observed below T_N in several antiferromagnetic Kondo systems, for instance YbNiAl and YbPtAl [31]. The $\rho(H)$ curve of YbCu_4Au in figure 4(a) is most likely to be indicating a spin-flop transition. In figure 4(b), the low-field regions of $\rho(H)$ for $x = 0.8$, 0.6, and 0.4 measured at 0.05 K are shown. Around $H = 0.3$ T, the $x = 0.8$ data show a shoulder, suggesting a spin-flop transition. On the other hand, no anomaly is observed for the $x = 0.6$ and 0.4 systems even at 0.05 K.

In figure 5, the $\rho(H)$ curves for $x = 0.2$ measured at 0.05 K and 1.5 K are shown. $\rho(H)$ at 1.5 K shows a maximum at $H_m = 4$ T. On the other hand, $\rho(H)$ at 0.05 K shows a minimum around $H = 1.5$ T, then begins to increase with field. The minimum around 1.5 T has been found to appear below 0.9 K and becomes more pronounced with decreasing temperature. This $\rho(H)$ behaviour cannot be attributed to the possibility of magnetic ordering below 0.9 K, because the temperature dependence of ρ for $x = 0.2$ shows no anomaly below 1.4 K. Instead,

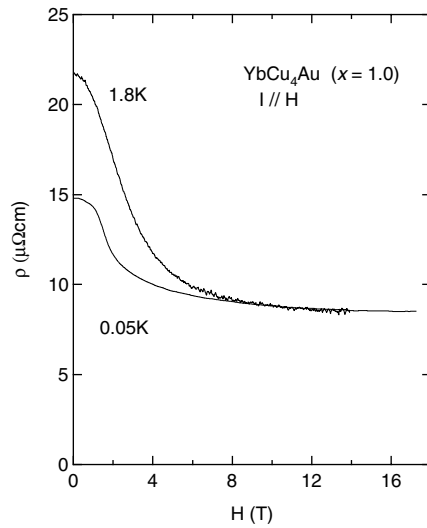


Figure 3. The field dependence of the resistivity of YbCu_4Au measured at 1.8 K and 0.05 K.

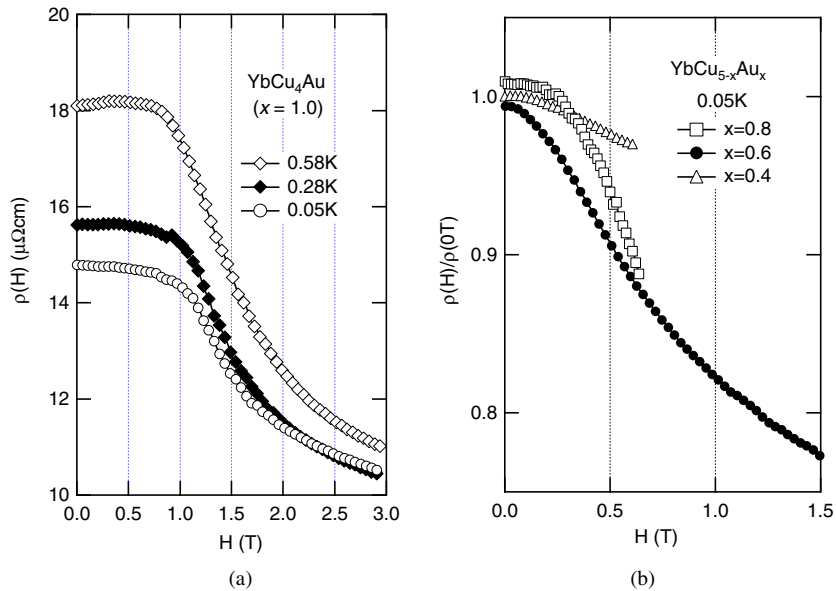


Figure 4. (a) The field dependence of the resistivity of YbCu_4Au measured at various temperatures. (b) The field dependence of the resistivity of $\text{YbCu}_{5-x}\text{Au}_x$ for $x = 0.8, 0.6,$ and 0.4 measured at 0.05 K. The data are divided by the values at $H = 0$ T and are slightly shifted for clarity.

a $-\ln T$ -dependent increase of $\rho(T)$ is observed for $x = 0.2$ in figure 1, which can be attributed to the impurity Kondo scattering. Impurity Kondo systems usually show negative magnetoresistance with increasing field [32]. We should note that the $\rho(H)$ for $x = 0.1$ also shows a subtle minimum around 0.5 T below 0.2 K, as is shown in the inset of figure 5. Hence, we can conclude that the minima in the $\rho(H)$ curves and the logarithmic increase of $\rho(T)$ are due to the impurity Kondo scattering caused by the substitution of Au for Cu.

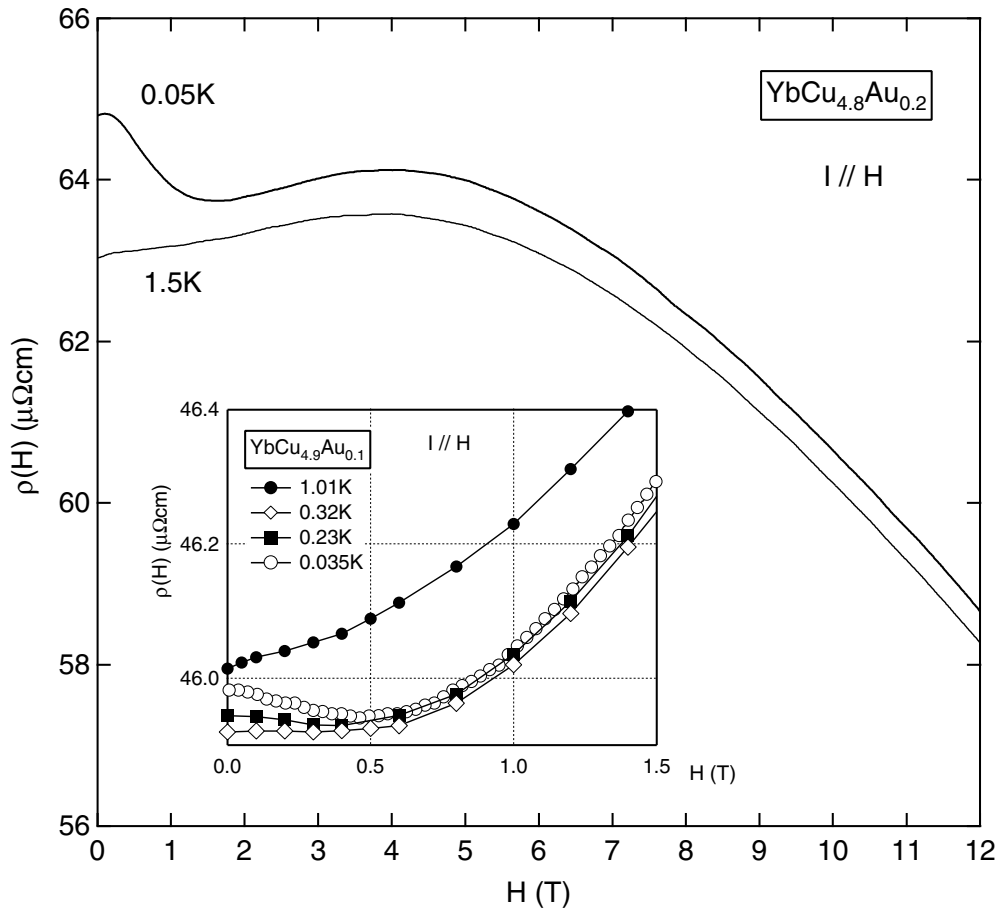


Figure 5. The field dependence of the resistivity of $\text{YbCu}_{4.8}\text{Au}_{0.2}$ measured at 0.05 K and 1.5 K. The inset shows the resistivity of $\text{YbCu}_{4.9}\text{Au}_{0.1}$.

3.3. The resistivity of $\text{YbCu}_{4.4}\text{Au}_{0.6}$ under magnetic fields

As was discussed before, the systems with $x = 0.4$ and 0.6 are expected to be in the vicinity of a magnetic instability, where enhanced magnetic fluctuations dominate the physical properties at low temperatures. Such magnetic fluctuations will be so much suppressed by magnetic fields that the ground state of the system may be changed by magnetic fields of several teslas. We have therefore measured the temperature dependence of ρ for $x = 0.4$ and 0.6 under magnetic fields. In figure 6, the $\rho(T)$ curves for $x = 0.6$ under fields of $H = 0, 0.5,$ and 1 T are shown. For $H = 0$ T, $\rho(T)$ has a maximum at $T_{max} = 0.4$ K and has an inflection point around $T = 0.1$ K, at which point the magnetic ordering is considered to set in. At $H = 0.5$ T, one finds that the maximum in the $\rho(T)$ curve becomes broad. The low-temperature region of $\rho(T)$ at $H = 0.5$ T is well fitted by the equation $\rho(T) = \rho_0 + CT^n$ with $\rho_0 = 45.88 \mu\Omega \text{ cm}$, $C = 9.746 \mu\Omega \text{ cm K}^{-1.38}$, and $n = 1.38$. The value of the exponent $n = 1.38$ is close to $3/2$. A $T^{3/2}$ -dependent $\rho(T)$ is observed in CeNi_2Ge_2 at low temperature, as has been discussed in terms of the quantum critical state with strong antiferromagnetic fluctuations [33]. Spin-fluctuation theory also predicts that a $T^{3/2}$ -dependence of $\rho(T)$ would

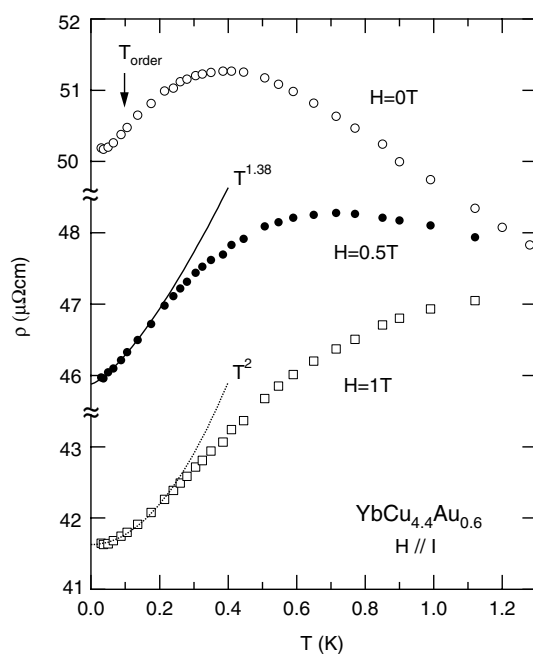


Figure 6. The temperature dependence of the resistivity of $\text{YbCu}_{4.4}\text{Au}_{0.6}$ measured in magnetic fields of $H = 0, 0.5,$ and 1 T. The solid curve indicates a $T^{1.38}$ -dependence. The dotted curve represents a T^2 -dependence.

appear in the antiferromagnetic critical state at low temperature [34]. Our results may suggest that the magnetic field has suppressed the intersite magnetic correlations in $\text{YbCu}_{4.4}\text{Au}_{0.6}$, reducing the magnetic ordering temperature, until the so-called non-Fermi-liquid state with strong spin fluctuations is evolved at $H = 0.5$ T. For $H = 1.0$ T, the maximum in $\rho(T)$ almost vanishes. The low-temperature region of $\rho(T)$ is well fitted by the equation $\rho(T) = \rho_0 + AT^2$, as is shown in figure 6. We have obtained $A = 14.24 \mu\Omega \text{ cm K}^{-2}$ from figure 6. A T^2 -dependence of the resistivity is a characteristic behaviour of the Fermi-liquid state. These results indicate that the intersite magnetic correlations in the $x = 0.6$ system are so reduced by the field that the Fermi-liquid state has recovered. This may be similar to the cases for $\text{CeCu}_{5.9}\text{Au}_{0.1}$ [35] and $\text{CeCu}_{4.8}\text{Ag}_{1.2}$ [36]. In the former case, the non-Fermi-liquid behaviour observed at zero field is replaced by a Fermi-liquid state at $H = 3$ T. In the latter case, the crossover from an antiferromagnetically ordered state at zero field, via non-Fermi-liquid behaviour around $H = 3$ T, to a Fermi-liquid state at $H = 5$ T is observed.

In a Fermi liquid, the value of A , the coefficient of the T^2 -term of the resistivity, is well known to be proportional to the square of γ , the electronic specific heat coefficient. Many heavy-fermion compounds appear to follow the relation $A/\gamma^2 = 1.0 \times 10^{-5} \mu\Omega \text{ cm mol}^2 \text{ K}^2 \text{ mJ}^{-2}$ [37, 38]. On the other hand, for the Kondo-lattice $\text{YbCu}_{5-x}\text{Ag}_x$ system, the values of A/γ^2 are almost $0.4 \times 10^{-6} \mu\Omega \text{ cm mol}^2 \text{ K}^2 \text{ mJ}^{-2}$ for the whole range of x [17]. Now, if we assume that the A/γ^2 value of $\text{YbCu}_{4.4}\text{Au}_{0.6}$ is the same as that of the $\text{YbCu}_{5-x}\text{Ag}_x$ systems, we can obtain the value $\gamma = 5 \text{ J mol}^{-1} \text{ K}^{-2}$ for $\text{YbCu}_{4.4}\text{Au}_{0.6}$ at $H = 1$ T. Thus, a quite massive electron state may be developed in $\text{YbCu}_{4.4}\text{Au}_{0.6}$ at low temperatures in a field of $H = 1$ T.

On the other hand, the $\rho(T)$ curve for $x = 0.4$ under a magnetic field is almost temperature independent, and a distinct change caused by fields has not been observed in this system.

4. Summary

We have investigated the temperature and the field dependence of ρ for $\text{YbCu}_{5-x}\text{Au}_x$. We have estimated the Néel temperature of YbCu_4Au to be 0.7 K; this value is in good agreement with those reported previously. We have found that the $x = 0.8$ and 0.6 systems each show a maximum in the $\rho(T)$ curve, which is most likely to be due to the onset of magnetic ordering. The magnetic ordering temperature, T_{order} , is estimated as $T_{order} = 0.4$ K and 0.1 K for $x = 0.8$ and 0.6, respectively. A large negative magnetoresistance has been observed in these systems, especially for $x = 0.8$. We have observed shoulders in the $\rho(H)$ curves for $x = 1.0$ and 0.8 measured at 0.05 K at 1.0 T and 0.3 T, respectively. The shoulder could be due to a spin-flop transition caused by the field.

For $x = 0.2, 0.1$, and 0, our results find no evidence of magnetic order. Thus we conclude that these systems remain paramagnetic at least down to 0.04 K ($x = 0.2$ and 0.1) or to 0.2 K ($x = 0$). The field dependence of ρ for $x = 0.2$ and 0.1 shows a maximum at H_m . This behaviour is considered to be a characteristic of a non-magnetic Kondo-lattice state. From the value of H_m , one finds that T_K for $\text{YbCu}_{5-x}\text{Au}_x$ decreases rapidly with the increase of x . This decrease of T_K cannot be explained by the chemical pressure effect. At lower temperatures, we have observed both a minimum and a maximum in the $\rho(H)$ curve, for $x = 0.1$ and 0.2. This can be understood as the consequence of coexistence of the impurity Kondo effect and the Kondo-lattice formation. This may be caused by the substitution of Au for Cu. For $x = 0.4$, the $\rho(H)$ curve no longer shows a maximum, but decreases linearly with increasing field. Thus, we can say that the Au substitution causes not only the decrease of T_K , but also the destruction of the Kondo-lattice state. This feature resembles the case for the $\text{YbCu}_4\text{Ag}_{1-x}\text{Au}_x$ system [25]. The wave function of Au may hybridize strongly with that of Yb, giving rise to a significant change in the local 4f electronic state. In addition to the crystal-field effect, this effect may have to be taken into account to explain the decrease of T_K with increasing x .

The value of T_{order} is expected to reach zero for an Au concentration of $x = 0.4$ –0.6. Hence, the so-called quantum critical state may evolve around these values of x . We have measured $\rho(T)$ for $x = 0.6$ in magnetic fields. We found that $\rho(T)$ at $H = 0.5$ T shows almost $T^{3/2}$ -dependence, which is predicted theoretically for the heavy-fermion systems with strong antiferromagnetic fluctuations. At $H = 1$ T, $\rho(T)$ was found to obey a T^2 -power law, characteristic of the Fermi-liquid state. Hence, this behaviour can be understood as the crossover caused by a magnetic field from the magnetically ordered state, via a non-Fermi-liquid state, to the Fermi-liquid state.

Acknowledgments

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