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Systematic composition dependence in $YbCu_{5-x}Au_x$ from dense Kondo to antiferromagnetic system through quantum critical point around $x=0.2–0.4$

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Abstract

Temperature and magnetic field variations of the electric resistivitiy (ρ) as well as temperature dependence of the magnetic susceptibility (χ) are reported for the $YbCu_{5-x}Au_x$ system with the C15b-type structure. These results indicate a systematic composition dependence from dense Kondo to RKKY antiferromagnetic system via quantum critical point with $x=0.2–0.4$. The logarithmic temperature dependence of ρ was observed in the middle temperature range for all the compositions, implying that the incoherent Kondo scattering is dominant in this T-region. With decreasing temperature, the ρ tends to saturate, makes a maximum, then decreases toward the Fermi liquid ground state due to the coherent dense Kondo effect ($x<0.2$), or toward the antiferromagnetic ground state due to the RKKY interaction (for $x>0.4$). The non-Fermi liquid behavior was observed around the quantum critical composition ($x=0.2–0.4$).

Keywords: Dense Kondo effect; Heavy Fermion; Fermi liquid; Non-Fermi liquid; $YbCu_5$; High pressure synthesis

1. Introduction

The cubic C15b ($AuBe_5$)-type intermetallic compounds based upon Yb and Cu exhibit various marvelous physical properties such as valence transition phenomenon ($YbCu_4In$) [1,2], dense Kondo behavior ($YbCu_4Ag$) [2–5] and antiferromagnetic ordering ($YbCu_4Au$ and $YbCu_4Pd$) [3–8]. Recently, the $YbCu_5$, which should be a key compound to understand the electronic properties of Yb in this system, has been successfully synthesized in a single phase of C15b-type under high pressure of 1.5 GPa at 900°C [9–12]. $YbCu_5$ has been found to show dense Kondo behavior having Fermi liquid ground state with very heavy effective mass of conduction electrons: the magnetic susceptibility (χ) has the maximum around $T_{max}\approx10$ K, the electric resistivity (ρ) follows T^2-dependence below T_{max}, and the electronic specific heat coefficient (γ) has a very large value of 550 mJ mol^{-1} K^{-2} [9–12].

The $YbCu_{5-x}T_x$ (T=Ag and In) system has given us valuable information on the various electronic state of Yb in this system [11–18]. In $YbCu_{5-x}Ag_x$, the systematic dense Kondo behavior was observed with the Kondo temperature T_K changing systematically from 80 to 50 K for decreasing x from 1 to 0 [12–14,17]. This behavior was successfully explained by the chemical pressure effect due to the difference of the ionic radii of Cu and Ag [12,13]. In $YbCu_{5-x}In_x$, the systematic change from the intermediate valence system with valence transition to the dense Kondo system was observed for decreasing x [11,12,15–17]. Therefore, the study on the $YbCu_{5-x}Au_x$ system between $YbCu_5$, the dense-Kondo heavy-Fermion compound with T_K of about 50 K [9–12], and $YbCu_4Au$, the RKKY antiferromagnetic compound with the Néel temperature T_N below 1 K [3–8], should give us supplementary information to the $YbCu_{5-x}T_x$ (T=Ag and In) system. Furthermore, since there exists a competition between the Kondo and the RKKY interactions in $YbCu_{5-x}Au_x$, this system should cross the critical region between the antiferromagnetically ordered and the dense Kondo nonmagnetic states with x. This region should be the so-called quantum critical point, in which many anomalous phenomena represented by keywords such as non-fermi liquid and non-BCS superconducting states can be expected to occur [19,20].

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2. Experimental

The samples of $YbCu_{5-x}Au_x$ were prepared from the mixture with nominal amounts of 99.9% pure ytterbium, 99.99% pure Cu and Au metals by Ar arc-melting, followed by annealing in evacuated quartz tubes at 750°C for 10 days. The samples of cubic $YbCu_{5-x}Au_x$ with $x=0$ and 0.1 thus prepared were treated under the pressure of 1.5 GPa at 900°C for 1 h by use of a piston-cylinder type high pressure apparatus. The phase identification of the samples was conducted by X-ray powder diffraction (XRPD), utilizing Cu Kα radiation. The metallographic texture was observed by scanning electron microscopy (SEM), and the chemical composition was determined by energy dispersive X-ray spectroscopy (EDS).

Magnetic susceptibility (χ) was measured by a superconducting quantum interference device (SQUID) magnetometer (Quantum Design, MPMS5). Electric resistivity (ρ) was measured by dc four-probe method. The ρ at very low temperatures and under magnetic fields was measured by utilizing ac four-probe method. The error of the absolute value of ρ would be within 20%, which are mainly coming from the size-estimation of the sample. The low temperature experiments in the temperature range of 0.03–1.5 K were conducted with a 3He–4He dilution refrigerator. Static magnetic fields up to 18 T were generated by a superconducting magnet for measuring the magnetic field dependence of ρ.

3. Results and discussion

The composition x dependence of the cubic lattice parameter (a) in the C15b-type $YbCu_{5-x}Au_x$ is presented in Fig. 1. As a whole, the values of a follow the Vegard's law (the straight line in the figure). However, a slightly positive deviation from Vegard's law was observed below $x=0.4$ in Fig. 1, which corresponds to the quantum critical region as will be mentioned later, and may be related to some physical properties in this region.

The temperature variations of the inverse magnetic susceptibility ($1/\chi$) of $YbCu_{5-x}Au_x$ are shown in Fig. 2. The composition dependence of Weiss temperature (θ) is also shown in the inset of Fig. 2. At high temperatures (>50 K), independently of x, χ follows a Curie–Weiss law behavior with the effective paramagnetic moment, μ_{eff}, of 4.3–4.5 μ_B/Yb, which corresponds well to the trivalent Yb-ion moment ($\mu_{eff}=4.54$ μ_B for $J=7/2$ and $g_J=8/7$), indicating that Yb is in a trivalent state through x. On the other hand, the value of θ changes from -27.2 K ($x=0$) to -5.5 K ($x=1.0$) with increasing x. Since the absolute value of θ is proportional to T_K, T_K in this system comes to be lower with increasing x. Finally, the RKKY interaction exceeds the Kondo interaction in $YbCu_4Au$ which was confirmed to show antiferromagnetic ordering at low temperatures below 1 K by various experiments including

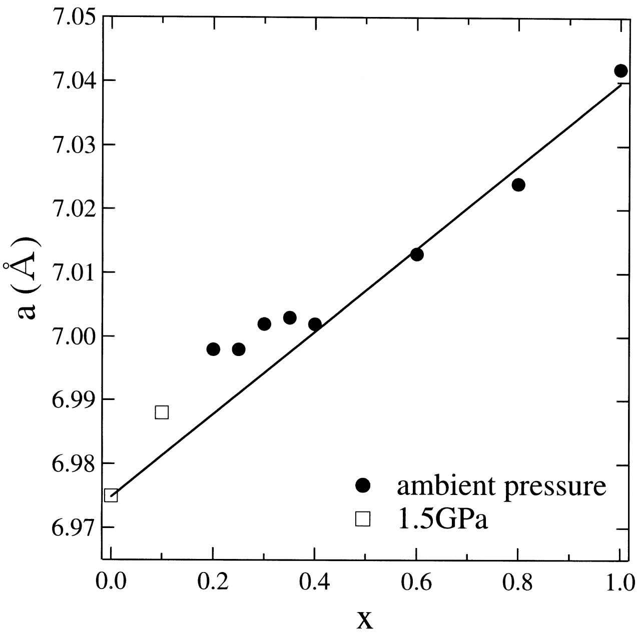

Fig. 1. Lattice parameter a (Å) vs. x of the cubic $YbCu_{5-x}Au_x$ with the C15b ($AuBe_5$-type) structure.

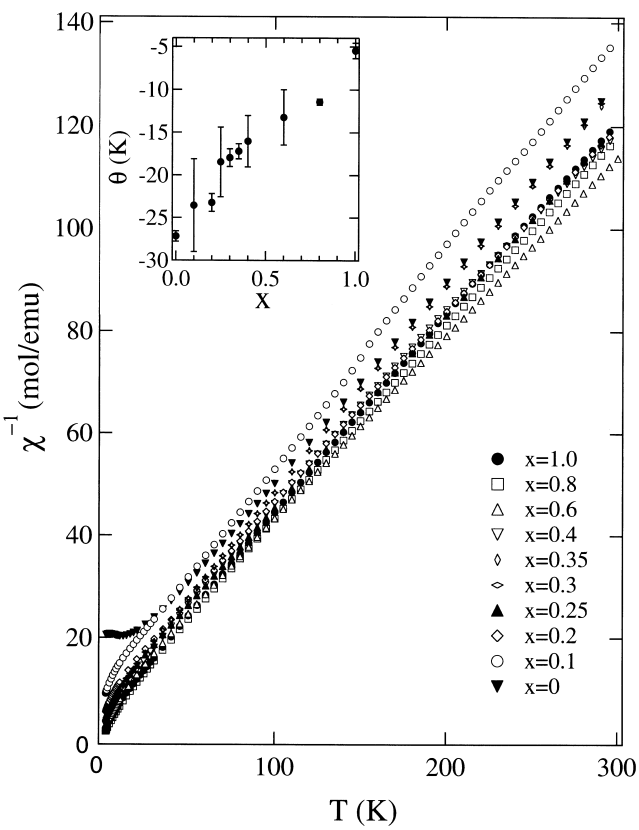

Fig. 2. Temperature dependence of inverse magnetic susceptibility ($1/\chi$) of the cubic $YbCu_{5-x}Au_x$. The inset shows the x-dependence of the Weiss temperature (θ).

neutron diffraction [3–8]. The χ of $YbCu_5$ shows broad maximum around T_{max}, and decreases toward the Pauli paramagnetic Fermi liquid state at low temperatures, resulting in the upward deviation of the $1/\chi$–T curve from the Curie–Weiss law below about T_{max} as seen in Fig. 2. For the other compositions, $1/\chi$ deviates downward from the Curie–Weiss law, which may be due to the crystal electric field effect: the degeneracy of the energy level of Yb 4f-electrons is solved into several levels in the presence of the crystal field. So, the degree of freedom is reduced by the crystal field effect. The ordered moment of $YbCu_4Au$ was obtained as very small value of 0.85 μ_B (at 0.04 K) [6] in compared with the paramagnetic moment, which should be concerned with this crystal field effect.

In Fig. 3, are presented the temperature variations of the electric resistivity (ρ) in the $YbCu_{5-x}Au_x$ system measured in the temperature region higher than about 1.5 K. The negative logarithmic temperature dependence ($-\log T$) of ρ was observed in the middle temperature range for all the compositions, implying that the incoherent Kondo effect is dominant in this T-region. With decreasing temperature, ρ for $x<0.2$ tends to saturate, makes a maximum, then decreases toward the fermi liquid ground state due to the coherent dense Kondo effect. The value of T_K can be estimated here as the temperature where ρ starts to deviate from the ($-\log T$)-dependence as shown in Fig. 3. T_K decreases with increasing x in agreement with the $1/\chi$–T data. At low temperatures, T^2-dependence of ρ was observed (see the inset of Fig. 3), indicating the Kondo lattice formation below the ρ–T maximum temperature, T_{max}. On the other hand, ρ for $x>0.4$ makes a maximum and actually again increases towards a second maximum and finally decreases toward the AF ordering for decreasing temperature. The non-Fermi liquid behavior was observed around the quantum critical composition ($x=$ 0.2–0.4): ρ is increasing and comes to be constant with decreasing temperature. The temperature variations of ρ at temperatures lower than 1.5 K are shown in Fig. 4a and b. For $x=1.0$, 0.8 and 0.6, ρ takes the second maximum around 1 K, then shows a decrease in decreasing temperature. Here, the value of $d\rho/dT$ takes a maximum below the ρ–T maximum temperature (see the inset of Fig. 4a). Let us suppose that this temperature is the Néel temperature T_N, T_N can be determined as 0.6, 0.4 and 0.2 K for $x=1.0$, 0.8 and 0.6, respectively, as shown in Fig. 4a. The value of T_N of 0.6 K for $x=1.0$ is very close to that determined by Rossel et al. [3]. Furthermore, the temperature variations of ρ were found to show the relation: $\rho=\rho_0+CT^n$ with $n=1.74$, 1.30 and 1.58 for $x=1.0$, 0.8 and 0.6, respectively. Those values of n is very close to $3/2$, which has been predicted theoretically by the SCR theory for the heavy Fermion system with strong antiferromagnetic spin fluctuations [21]. The deviation temperature from this $T^{3/2}$ relation was found to coincide with T_N just determined above as seen in Fig. 4a, which may indicate the validity of the present determination of T_N. For $x=0.0$ and 0.1, ρ becomes to have constant value of ρ_0, the residual resistivity at low temperature as shown in Fig. 4b. Near the critical region with $x=0.2$ and 0.4, ρ increases but tends to be saturated to a constant value with decreasing temperature, which may be concerned with quantum critical behaviors.

Magnetic field variations of the electric resistivity measured at 0.05 K with the electric current applied parallel to the magnetic field ($\rho_{//H}$) are shown in Fig. 5. The $\rho_{//H}$ of $YbCu_5$ increases monotonically with fields up to 17 T, while those of $YbCu_{4.9}Au_{0.1}$ and $YbCu_{4.8}Au_{0.2}$ indicate the maxima around 8 and 4 T, respectively. The positive magnetoresistance seems to be common in dense Kondo systems with Fermi liquid ground state [22–24]. The positive magnetoresistance can be qualitatively explained in terms of the change of the 4f electronic state by magnetic fields from the itinerant 4f electronic state without local moment having low resistance at low magnetic fields to the localized 4f state at high fields with magnetic moment scattering conduction electrons, giving rise to high resistance at high fields. This type of crossover

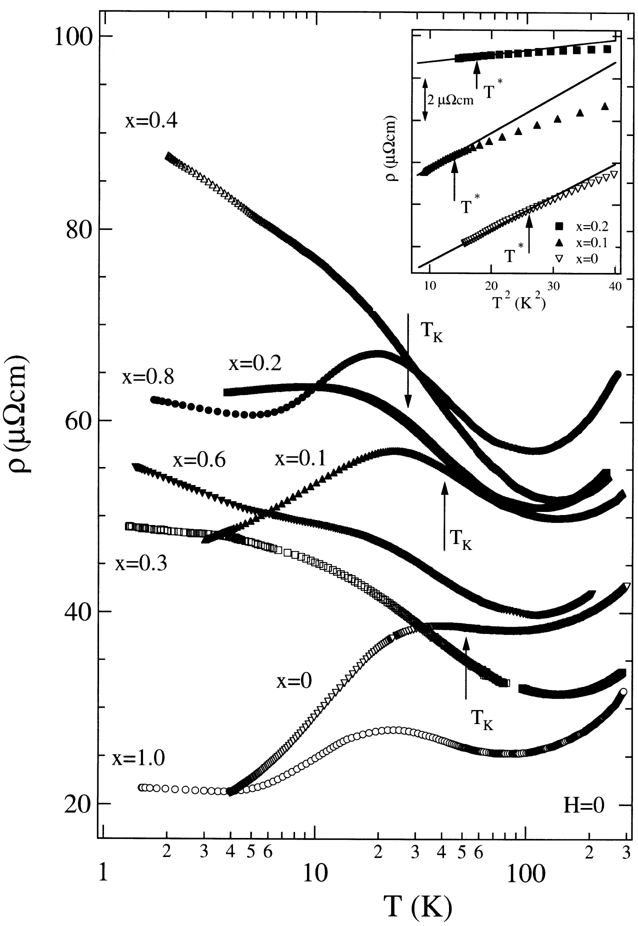

Fig. 3. Temperature dependence of electric resistivity (ρ) of the cubic $YbCu_{5-x}Au_x$ measured above 1 K. In the inset, ρ is plotted against T^2 for $x=0$, 0.1 and 0.2. The value of ρ for each sample is offset along with each other for describing all the data in the same inset figure. ρ follows T^2-dependence below T^*.

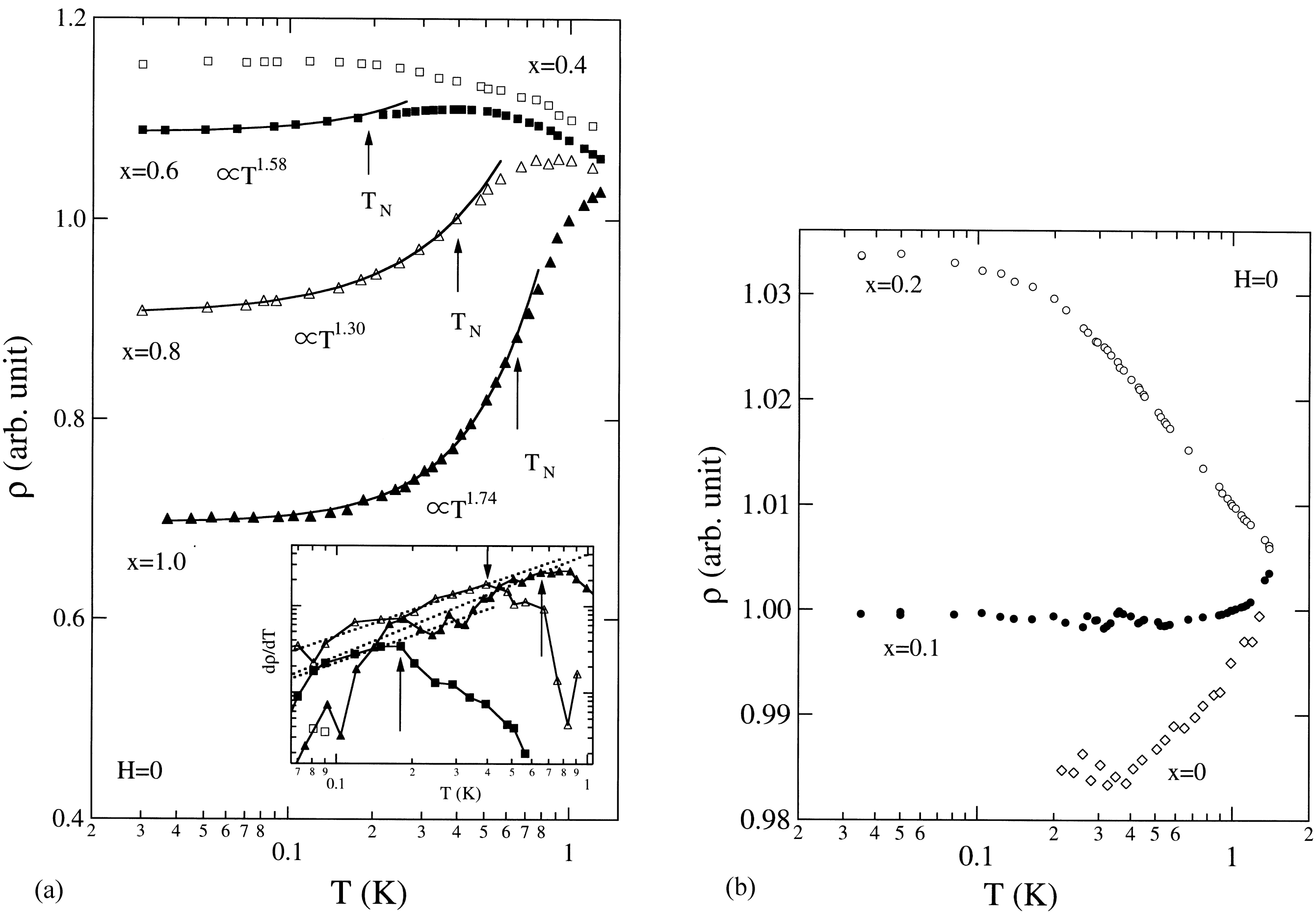

Fig. 4. (a) Temperature dependence of electric resistivity (ρ) of the cubic $YbCu_{5-x}Au_x$ measured below 1.5 K for $x=1.0$, 0.8, 0.6 and 0.4. The solid curves represent the power-law relation: $\rho=\rho_0+CT^n$ with $n=1.74$, 1.30 and 1.58 for $x=1.0$, 0.8 and 0.6, respectively. The temperature dependence of $d\rho/dT$ is shown in the inset in the form of log–log plots. The dashed lines stand for $d\rho/dT=nCT^{n-1}$. Above T_N, ρ deviates from this power law behavior. (b) Temperature dependence of electric resistivity (ρ) of the cubic $YbCu_{5-x}Au_x$ measured below 1.5 K for $x=0.2$, 0.1 and 0.0.

of the 4f electronic state was observed in the high-field magnetization measurements for $YbCu_4Ag$ [2] and $YbCu_5$ [9] by using the pulsed magnetic field up to 40 T. Theoretically, the maximum in ρ has been predicted to take place at the metamagnetic transition field (H_m) by the calculation based upon the periodic Anderson model [25]. The magnetic fields at the maximum ρ of 8 and 4 T for $x=0.1$ and 0.2 are corresponding to H_m of these two compounds. For $YbCu_5$, the magnetic field (17 T) is smaller than H_m of this compound. For $x=0.4$, $\rho_{//H}$ decreases rapidly with increasing H. The large negative magnetoresistance is considered to be observed in the antiferromagnetic Kondo system around T_N. The large drop in $\rho_{//H}$ for $x=0.6$, especially, should correspond to the spin flop field, above which $\rho_{//H}$ tends to be constant quickly with the saturation of Yb moment by fields as seen in Fig. 5. The field dependence of $\rho_{//H}$ was measured at 0.05 and 1.8 K for $YbCu_5$ as shown by the absolute values in the inset of Fig. 5. At high magnetic field above 9 T where the magnetization is saturated and, thus, the magnetic moments lay almost parallel to the field, the values of $\rho_{//H}$ at 0.05 and 1.8 K show good agreement with each other and take about 9 μΩ cm, which should be a residual resistivity without any magnetic contributions. Therefore, the magnetoresistance can also be explained by the systematic change of the Yb 4f-electronic state from the nonmagnetic dense Kondo to antiferromagnetic Kondo system with increasing x.

4. Conclusion

Temperature and magnetic field variations of the electric resistivitiy (ρ) as well as temperature dependence of the magnetic susceptibility (χ) in $YbCu_{5-x}Au_x$ with the C15b-type a structure indicate a systematic composition dependence from the itinerant nonmagnetic dense Kondo system to the RKKY antiferromagnetic system. From the present experiments, we can obtain the electronic phase diagram shown in Fig. 6, where the various characteristic temperature obtained from the experiments are presented: T_K is the Kondo temperature, T_{max} the maximum temperature at the

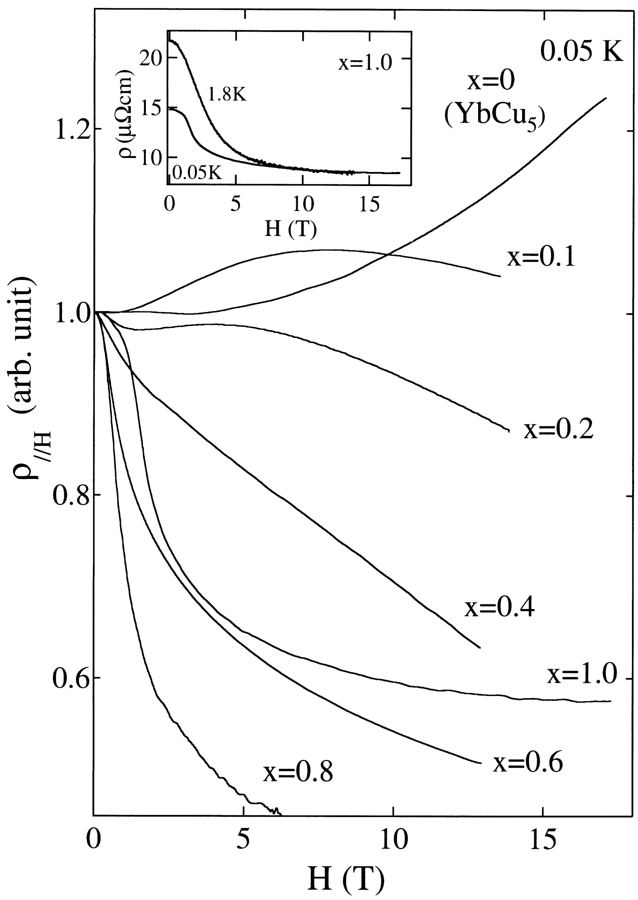

Fig. 5. Magnetic field dependence of electric resistivity (ρ) (magneto-resistivity) of the cubic $YbCu_{5-x}Au_x$ measured at 0.05 K. The inset indicates the data for $x=1.0$ measured at 0.05 K (below T_N) and at 1.8 K (above T_N). The current was applied parallel to the magnetic field in the experiments.

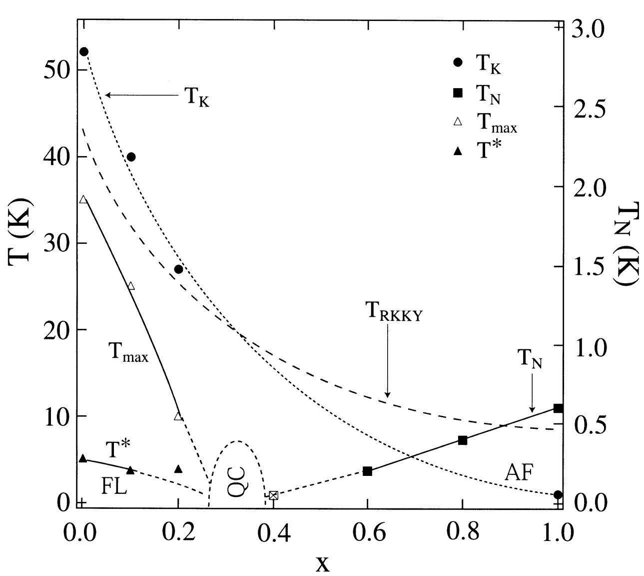

Fig. 6. Electronic phase diagram of the cubic $YbCu_{5-x}Au_x$. Here, T_K represents the Kondo temperature, T_{max} the maximum temperature at the ρ–T curve, T^* the temperature below which ρ follows T^2-dependence, T_N the Néel temperature and T_{RKKY} the temperature scale indicating the strength of the RKKY interaction. FL stands for the Fermi liquid state; AF, the antiferromagnetic state, and QC, the quantum critical region.

ρ–T curve, T^* the temperature below which ρ follows T^2-dependence, T_N the Néel temperature and T_{RKKY} the temperature scale indicating the strength of the RKKY interaction. The quantum critical region (QCR) is realized in the vicinity of $0.2<x<0.4$ between the Fermi liquid (FL) nonmagnetic state and the antiferromagnetic (AF) Kondo state. The non-fermi liquid behavior was observed around the quantum critical composition ($x=0.2$–0.4).

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