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Effects of alloying in the $Ce(Cu_{1-x}Au_x)_6$ ($x \leqslant 0.25$) system

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Abstract. We present x-ray diffraction, electrical resistivity, thermopower, magnetic susceptibility and specific heat measurements on a series of $Ce(Cu_{1-x}Au_x)_6$ compounds with $x \leqslant 0.25$. These measurements indicate that alloying produces an increase in cell volume and changes in the sd density of states. Thus, while $CeCu_6$ has a non-magnetic ground state, addition of Au leads to the onset of magnetic order (e.g. $T_N \simeq 2$ K for the Au alloy with $x = 0.15$). The results also reveal alloying with Au produces modifications in the crystalline electric field scheme.

1. Introduction

$CeCu_6$ is a heavy fermion system which exhibits neither superconductivity nor magnetic order down to 20 mK [1–3]. At high temperature the properties of pure $CeCu_6$ indicate the presence of localised Ce 4f moments. These moments produce a Curie–Weiss like susceptibility and act as individual Kondo scattering centres producing a region in which the magnetic resistivity exhibits a logarithmic temperature dependence.

At lower temperatures the properties of $CeCu_6$ are dominated by two competing interactions.

(i) The oscillatory RKKY interaction between Ce 4f moments which is characterised by the temperature $T_{RKKY} \sim J^2N(E_F)$, where $N(E_F)$ is the density of states at the Fermi level and $J(<0)$ is the coupling constant between the 4f shell and the conduction band. Low temperature neutron scattering data [4, 5] indicates the presence of antiferromagnetic correlations in pure $CeCu_6$.

(ii) The screening of the 4f moments by conduction electrons known as Kondo compensation characterised by the Kondo temperature $T_K \approx T_F \exp(-1/JN(E_F))$. In the case of $CeCu_6$ this screening leads to a non-magnetic ground state. In many heavy fermion systems a suitable change in J and/or $N(E_F)$ due to variations in the external pressure or alloying can produce a change in the nature of the ground state, e.g. the $Ce(Cu_{1-x}Al_x)_5$ system [6]. This paper examines the effects on the properties of $CeCu_6$ of replacing Cu with isoelectric Au. Since Au has a larger ionic radius than Cu one expects to observe an increase in the cell volume, leading to a decrease in the magnitude

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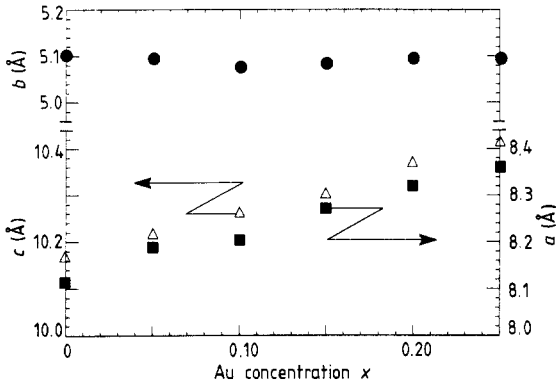

Figure 1. The lattice parameters (a (■), b (●) and c (△)) versus Au concentration x, for $Ce(Cu_{1-x}Au_x)_6$ where $x \leqslant 0.25$.

of J, which in turn may lead to the onset of magnetic order. Adding Au may also produce changes in the nature of the conduction electrons.

2. Experimental details

$CeCu_6$ has an orthorhombic structure with values for the lattice parameters given in figure 1. $CeAu_6$ exists [7] although there is no report in the literature of its structure. Samples of $Ce(Cu_{1-x}Au_x)_6$ with $x \leqslant 0.25$ were prepared by arc melting under an argon atmosphere. All the samples were remelted several times to ensure homogeneity and annealed in evacuated quartz tubes for 7 days at 700 °C. Metallographic specimens were prepared from the annealed samples. All the samples containing less than 20% Au were found to be essentially single phase with only small traces of Ce oxides present. Samples containing greater than 20% Au showed signs of a small amount of a second phase. Examining the phase diagram of the Ce–Cu system [8] it seems likely that this is a little free copper. X-ray powder diffraction patterns were obtained at room temperature from the annealed samples. The x-ray patterns all have the orthorhombic structure of the parent compound $CeCu_6$. Resistivity measurements were performed using a conventional four-probe technique. Thermopower measurements were made by a differential method using Pb as a reference material. AC susceptibility measurements were made using a mutual inductance technique. Specific heat measurements in the temperature range 1.5–30 K were carried out in a fully automated Nernst calorimeter.

3. Results

The lattice parameters of $Ce(Cu_{1-x}Au_x)_6$ for $x \leqslant 0.25$ are shown in figure 1. The values for pure $CeCu_6$ are in close agreement with the published data [9]. Addition of Au produces an increase in the values of a and c, whilst the values for b remain almost constant throughout the sample series. The increases in a and c are more rapid than those previously reported for samples with Au concentrations of up to 10% [10]. These changes result in a continuous increase in the cell volume with increasing Au concentration.

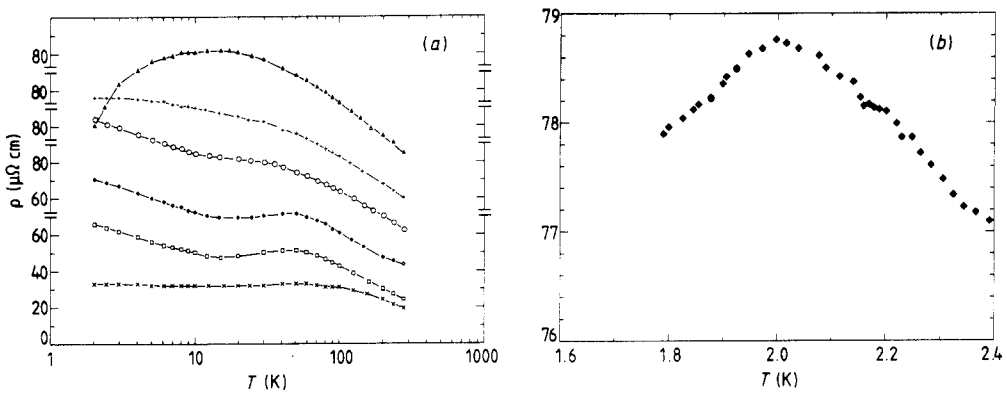

Figure 2. (*a*) The temperature dependence of the magnetic component of the electrical resistivity of selected $Ce(Cu_{1-x}Au_x)_6$ alloys given by $\rho(Ce(Cu_{1-x}Au_x)_6) - \rho(La(Cu_{1-x}Au_x)_6)$ in a semi-logarithmic representation. Here $x = 0$ (△), 0.05 (+), 0.10 (○), 0.15 (◇), 0.20 (□) and 0.25 (×). (*b*) The low temperature maximum in the resistivity of the alloy $Ce(Cu_{0.85}Au_{0.15})_6$ shown over a narrower temperature range.

Table 1. Some properties of selected $Ce(Cu_{1-x}Au_x)_6$ alloys taken from transport data.

x	0.00	0.05	0.10	0.15	0.175	0.20	0.25
Low T_{max} (K)	15	≤1.5	—	2.0	2.05	—	—
High T_{max} (K)	—	30.0	35.0	39.0	44.5	43.5	49.0
$\rho_{273\,K}$ ($\mu\Omega$ cm)	59.5	61.8	61.0	60.9	58.9	63.0	58.4
$-d\rho_{magnetic}/d \ln T$	25.5	22.5	20.5	19.9	19.5	18.9	11.4
$S(T)_{max}$ (K)	50.0	—	86.0	—	—	80.0	—

Figure 2 and table 1 contain details of the behaviour of the electrical resistivity of $Ce(Cu_{1-x}Au_x)_6$ alloys for $x \leq 0.25$. The temperature dependence of the magnetic component of the electrical resistivity is given by $\rho(Ce(Cu_{1-x}Au_x)_6) - \rho(La(Cu_{1-x}Au_x)_6)$ where $La(Cu_{1-x}Au_x)_6$ alloys were used as non-magnetic, isostructural reference compounds. Several features in the resistivity data must be highlighted. The value of the total resistivity at high temperature is almost independent of Au concentration. This can be understood since Au substitution is not expected to lead to a large increase in the residual resistivity. At the same time the magnitude of the magnetic resistivity at high temperature is dominated by the incoherent Kondo scattering of the Ce sublattice.

The temperature T_M at which a maximum is seen in the resistivity (= 15 K for pure $CeCu_6$) is rapidly suppressed as Au is added. For 2% Au, $T_M = 6$ K whilst it has been suppressed below 1.5 K for 5% Au. This behaviour contrasts with the effects of replacing Ce with Y, La or Gd where much larger concentrations of dopant (~20%) are required to suppress T_M below 1.5 K [11]. This maximum in $\rho(T)$ is associated with the onset of coherence in $CeCu_6$ i.e. the periodic array of Ce atoms scatters coherently leading to a decrease in the resistivity. The rapid reduction in T_M when alloying on the Cu sites with either Au [11], Ag [12], Al or Ni [11, 13] suggests that this maximum is not a crystal field effect (as it probably is in part in the $CeAl_3$ system) and that the onset of coherence

which leads to the appearance of the maximum in the resistivity is sensitive to even small changes in the nature of the conduction band electrons. As the Au concentration is increased a feature appears in the data around 30 to 50 K. The temperature at which this maximum occurs increases with increasing Au concentration. This feature separates two regions where the resistivity has a logarithmic temperature dependence. The slope $d\rho_{magnetic}/d \ln T$ at high temperature decreases with increasing Au concentration. Cornut and Coqblin [14] have developed a model which considers the form of the magnetic component of the resistivity in the presence of Kondo and crystal field effects. In their model the magnetic contribution to the resistivity has a logarithmic temperature dependence in both a high and a low temperature region separated by a peak around the crystal field splitting temperature. Hanzawa *et al* [15] derived a relationship between the true Kondo temperature T_K and T_K^H. T_K^H is the apparent Kondo temperature of a system at temperatures greater than the crystal field splittings. They showed that $T_K^H = (T_K\Delta_1\Delta_2)^{1/3}$ where Δ_1 and Δ_2 are the crystal field splittings between the ground state doublet and the first excited level and the first and second excited doublets respectively. It is around T_K^H that a peak in the resistivity due to a combination of the Kondo effect and the crystal field splitting is expected to be observed. In the case of pure $CeCu_6$ the 4f level is split into three doublets by the crystal field. The crystal field splitting energies Δ_1 and Δ_2 and the true Kondo temperature have been calculated from experimental data by several groups. Using specific heat data Fujita *et al* [16] estimated $\Delta_1 = \Delta_2 = 65$ K with $T_K = 3.9$ K. A fit to magnetic susceptibility data by Takayanagi *et al* [17] produced $\Delta_1 = 87$ K and $\Delta_2 = 210$ K, whilst Walter *et al* [18] used neutron data to calculate $\Delta_1 = 64$ K and $\Delta_2 = 128$ K. Penney *et al* [19] have presented magnetoresistance data which indicate that $T_K \simeq 2$–3 K. Substituting these values into $T_K^H = (T_K\Delta_1\Delta_2)^{1/3}$ gives values of T_K^H anywhere between 20 and 40 K. It has been argued that T_K^H and the temperature at which the resistivity of $CeCu_6$ begins to fall, due to the appearance of coherence, are too close together for any peak in the resistivity resulting from a combination of crystal field and Kondo effects to be resolved from the low temperature maximum which signals the onset of coherence.

This produces the single peak structure observed in $\rho(T)$ of $CeCu_6$. Addition of Au leads to a suppression in the low-temperature maximum associated with the onset of coherence. This should allow the observation of any feature due to a combination of the Kondo effect and the crystal field splitting which may have been masked by this peak. However, the effect of Au substitution on the crystal field splitting must also be considered. Thermal expansion data presented by Oomi *et al* [20] and de Visser *et al* [21] have indicated that Δ_1 and Δ_2 are expected to increase with pressure. Thus alloying with Au, which produces negative chemical pressure should reduce Δ_1 and Δ_2 suppressing the position of the maximum in the resistivity expected due to a combination of the Kondo effect and the crystal field splitting. However alloying also introduces distortions within the crystal lattice. In several systems (e.g. $Ce_{1-x}M_xAl_3$ where M is Y, Th or La [22]) these distortions can lead to rapid modifications of the crystal field scheme. It is suggested that a combination of a suppression of the peak in $\rho(T)$ attributed to the onset of coherence and an increase in the crystal field splitting, driven by a combination of crystallographic distortion and modifications in the nature of the conduction band electrons, produce conditions where a well defined peak in the resistivity due to the Kondo crystal field interaction becomes visible. The shift to higher temperature of this peak with increasing Au concentration indicates a continued increase in the crystal field splitting. For example, for $x = 0.20$, $T_K^H = 44$ K. Taking $T_K = 2$ K and $\Delta_1 = \Delta_2$ indicates crystal field splitting energies of 200 K. Cornut and Coqblin also showed that the slopes

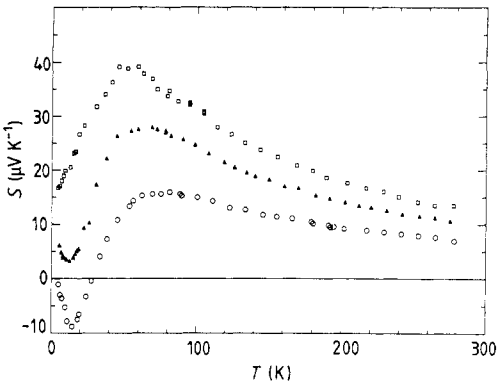

Figure 3. The temperature dependence of the thermopower S of some $Ce(Cu_{1-x}Au_x)_6$ alloys where $x = 0.00$ (□), 0.10 (▲) and 0.20 (○).

$d\rho_{magnetic}/d \ln T$ in the regions at both high and low temperature where the resistivity exhibits logarithmic temperature dependence, are related to the product $J^3N(E_F)$. The reduction observed in the magnitude of $d\rho_{magnetic}/d \ln T$ at high temperature with increasing Au concentration indicates that this product decreases as Au is added, and that for higher Au concentrations the Kondo interaction is considerably weakened.

At concentrations of $0.15 \leqslant x \leqslant 0.175$ a maximum in the resistivity is seen at approximately 2 K. It seems likely that this maximum signals the onset of long range magnetic order. This magnetic ordering is discussed more fully below.

The temperature dependence of the thermopower $S(T)$, of some $Ce(Cu_{1-x}Au_x)_6$ alloys are shown in figure 3. In common with many other non-magnetic heavy fermions ($CeAl_3$ [23], $CeCu_3Al_2$ [6]) the thermopower of $CeCu_6$ has extrema with unusually large absolute values. A large positive maximum with a peak value of 40 $\mu V\ K^{-1}$ is seen in $S(T)$ at 50 K. The addition of 10% Au reduces the magnitude of the maximum seen in $S(T)$ to 27 $\mu V\ K^{-1}$, whilst the position of the positive peak is shifted to 68 K [6]. The exchange of 20% of Cu by Au yields a further reduction in the magnitude of the thermopower maximum to about 16 $\mu V\ K^{-1}$ and to a shift of this positive peak to 80 K. Maekawa *et al* [24] calculated the temperature dependence of the thermopower $S(T)$ of Ce based heavy fermions. A large positive peak in $S(T)$ is attributed to an interplay between incoherent Kondo scattering and the crystal field and is in agreement with the resistivity data presented above. According to Bhattacharjee and Coqblin [25] the position of the maximum in $S(T)$ gives a measure of the overall crystal field splitting. Within this framework, the observed shift of the maximum in $S(T)$ from about 50 K in pure $CeCu_6$ to 80 K for the sample containing 20% Au again indicates a remarkable enlargement of the level splitting of nearly 60%. Since an increase in the volume of the unit cell usually leads to a reduction in the crystal field splitting, the experimentally observed increase of Δ_1 and Δ_2 points to a drastic reformation of the conduction electron band caused by the substitution of Cu by Au. Furthermore, the reduction of the $S(T)$ values and the formation of a minimum in $S(T)$ resulting from the increasing Au concentration is indicative of the diminishing strength of the Kondo interaction as long range magnetic order is established (see below). It is interesting to note that similar behaviour in $S(T)$ has been observed in the crossover from the magnetically ordered Kondo compound $CeCu_5$ to the non-magnetic heavy fermion compounds $CeCu_3Al_2$ and $CeCu_3Ga_2$ [26]. While $S(T)$ of the former compounds shows a pronounced minimum at low temperature and a flat maximum at elevated temperatures, the latter compounds

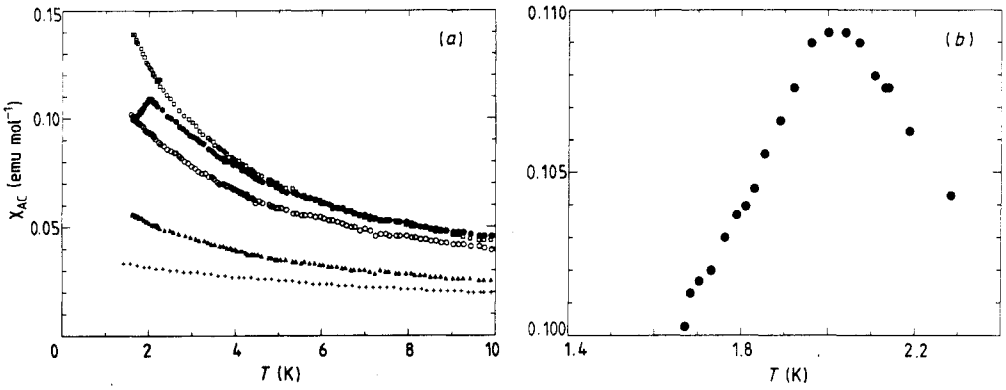

Figure 4. (*a*) The temperature dependence of the magnetic susceptibility of selected $Ce(Cu_{1-x}Au_x)_6$ alloys, where $x = 0$ (+), 0.05 (▲), 0.10 (○), 0.15 (●) and 0.20 (□). (*b*) The cusp in the susceptibility of the $Ce(Cu_{0.85}Au_{0.15})_6$ sample shown over a narrower temperature range.

Table 2. Some magnetic properties of selected $Ce(Cu_{1-x}Au_x)_6$ compounds.

x	0.00	0.05	0.10	0.15	0.175	0.20	0.25
θ_P (K)	−20.1	−18.2	−13.0	−8.4	−8.1	−7.1	−6.5
μ_{eff} (μ_B)	2.31	2.38	2.54	2.65	2.57	2.63	2.65
χ_{AC} (emu mol)$^{-1}$ at 4.2 K	0.029	0.038	0.064	0.076	0.063	0.078	0.064
T_{max} (K)	—	—	—	1.93	2.06	—	

exhibit a single positive maximum, with very large absolute thermopower values. Within this phenomenological trend it seems it may be possible that the interpretation of the observed $S(T)$ data can help distinguish between a long range magnetically ordered state and that of a ground state which is probably non-magnetic.

Figure 4 and table 2 contain details of the behaviour of the magnetic susceptibility for $Ce(Cu_{1-x}Au_x)_6$ alloys with $x \leq 0.25$. Plots of χ^{-1} against T reveal that all the alloys show Curie–Weiss like behaviour at high temperature. This is much as expected, since at high temperature the properties of these alloys are largely determined by the nature of the localised Ce ions. Fits to $\chi^{-1} = C/(T - \theta)$ give values for the effective moment and paramagnetic Curie temperature. The value of μ_{eff} for pure $CeCu_6$ is in close agreement with the published data and is independent of Au concentration. The value for $|\theta|$ for pure $CeCu_6$ is somewhat smaller than the values observed by other workers. The value of $|\theta|$ decreases with increasing Au concentration. Initially the substitution of Au leads to a rapid increase in the magnitude of χ at 4.2 K. At higher Au concentrations the value of χ at 4.2 K changes little with Au content. These results may indicate that T_K decreases with increasing Au concentration. Using the single ion Kondo model gives $T_K \approx |\theta|/4$ [27]. This analysis suggests that T_K decreases from 5 K for $x = 0$ to 1.8 K for $x = 0.2$ in the $Ce(Cu_{1-x}Au_x)_6$ system. One can also relate T_K to the absolute value of χ at low temperature, $T_K \simeq 0.15/\chi$ ($T \rightarrow 0$ K) [28]. The increase in χ at $T = 1.5$ K with Au concentration then suggests a T_K of approximately 4 K for pure $CeCu_6$ decreasing to

Table 3. Some properties of selected $Ce(Cu_{1-x}Au_x)_6$ alloys extracted from specific heat data.

x	0.00	0.03	0.083	0.13	0.15	0.175	0.20
γ (mJ mol^{-1} K^{-2})	235	—	145†	95	60	47	46
θ_D (K)	228.5	—	217.6†	206.5	206.3	201.2	196.9
T_N (K)	0.00	0.25†	0.55†	1.8	2.1	2.1	1.7

† Values taken from [29].

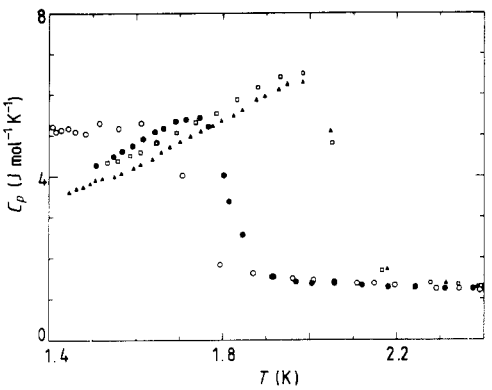

Figure 5. The specific heat plotted as a function of temperature for some $Ce(Cu_{1-x}Au_x)_6$ alloys where $x = 0.13$ (●), 0.15 (▲), 0.175 (□) and 0.20 (○).

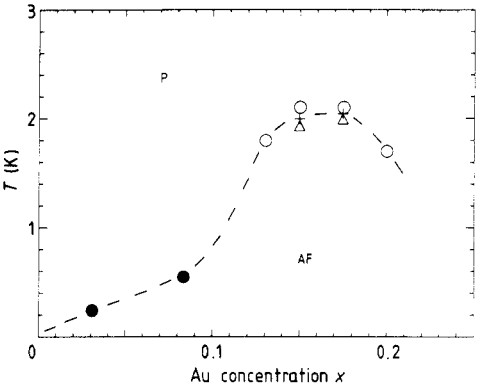

Figure 6. The proposed magnetic phase diagram for the $Ce(Cu_{1-x}Au_x)_6$ system for $x \leq 0.25$. Transition temperatures have been determined from specific heat (○), electrical resistivity (+) and magnetic susceptibility (△) measurements and from the specific heat data reported by Germann *et al* (●) in [29]. P and AF indicate the paramagnetic and antiferromagnetic regimes respectively.

around 1 K for $x = 0.2$. Below 40 K, $\chi(T)$ deviates from Curie–Weiss behaviour. For $0.15 \leq x \leq 0.175$ a peak appears in the susceptibility at approximately 2 K indicating a transition into a magnetically ordered state. No maximum is present above 1.5 K outside this concentration range.

Specific heat measurements were performed on $Ce(Cu_{1-x}Au_x)_6$ for $0.13 \leq x \leq 0.20$, between 1.5 and approximately 20 K. Between 10 and 20 K or the highest temperature reached in the experiment the specific heat can be described by $C = \gamma T + \beta T^3$ where γ is proportional to the electronic density of states and β can be related to θ_D, the Debye temperature. Values for γ and θ_D for the samples studied in this work and from two samples with lower Au concentration studied by Germann *et al* [29] are given in table 3. The steady decrease in θ_D is attributed to the higher Au mass. A decrease is observed in the value of γ, from 234 mJ mol^{-1} K^{-2} for pure $CeCu_6$ to just 46 mJ mol K^{-2} for $CeCu_6$ with 20% Au. It is not possible to categorically state that this reduction in γ indicates that the heavy fermion behaviour has been removed. This is because γ is temperature dependent and an upturn in C/T may occur at a lower temperature. Figure 5 shows the low temperature specific heat C plotted as a function of temperature. In all four samples measured, a sharp anomaly extending over only 0.2 K is observed. For $x = 0.13$, 0.15

and 0.175 the temperature of this anomaly (T_N) is calculated as the mid-point of the upturn in C. Only a rough estimate of the temperature of the anomaly can be made for the $x = 0.20$ sample, since it occurs at around 1.7 K, close to the lowest temperature which can be reached in the experimental apparatus.

4. Discussion

Because of the different functional dependence of T_{RKKY} and T_K on the values of J and $N(E_F)$, the reduction in the product $JN(E_F)$ indicated by the apparent decrease in T_K is expected to weaken the RKKY interaction less rapidly than the Kondo interaction, increasing the tendency toward magnetic order. This expectation is confirmed, since, as well as an increase in the absolute value of χ_{AC} a cusp is seen in the low temperature data for $0.15 \leqslant x \leqslant 0.175$. The cusp suggests the onset of magnetic ordering. The magnitude of the susceptibility at the peak seems to rule out ferromagnetic order. The cusp could be indicative of a spin-glass or antiferromagnetic order. The peak in the resistivity and the sharp anomaly in the specific heat point to long range antiferromagnetic order. The transition temperatures T_N extracted from the positions of peaks in the resistivity and susceptibility data and the mid-points of specific heat anomalies allow the construction of the magnetic phase diagram shown in figure 6. It can be seen from this diagram that there is a steady increase in the transition temperature with increasing Au concentration up to 15% Au. For higher Au concentrations the value of T_N saturates and then begins to fall (similar behaviour has been noted in the $Ce(Cu_{1-x}Ag_x)_6$ system [30]). For example, no peak is visible in the resistivity or susceptibility data of the 20% Au sample down to the lowest temperature available in these experiments. The onset of a transition is just discernible in the specific heat data. If the rapid reduction in T_N seen between 17.5% and 20% Au is continued to higher Au concentrations, the transition temperature would be reduced to 0 K around 30% Au. This decrease in the ordering temperature reflects the continued changes in J and $N(E_F)$ with increasing Au concentration, which produce reductions in the strength of both RKKY and Kondo interactions and lead eventually to a system in which both magnetic and Kondo interactions are considerably weakened.

Recently Germann and Löhneysen [31], have shown via measurements of the pressure dependence of T_N that the principle mechanism responsible for establishing magnetic order is the increase in cell volume produced by replacing Cu with Au. This expansion results in a decrease in J which in turn favours the stability of magnetic moments present within the system. However they also showed that changes in the band structure of the sd conduction electron bands due to alloying plays an important secondary role.

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