Influence of substitutions on the physical properties of $CeInCu_2$

Influence of substitutions on the physical properties of CeInCu₂

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Abstract. We present x-ray diffraction and electrical resistivity measurements on two series of compounds: $CeIn_{1-x}M_xCu_2$ (M = Al, Ga and Sn) and $CeIn(Cu_{1-x}T_x)_2$ (T = Fe, Ni, Pd and Pt) with $x \leqslant 0.2$. The change in electrical resistivity with alloying is discussed in terms of the effect of this on the Kondo interaction and crystalline electric field splitting. These are influenced by two different mechanisms: the unit-cell volume and the number of conduction electrons of the systems.

1. Introduction

CeInCu₂ crystallizes with the Heusler $L2_1$ type of cubic structure [1]. It exhibits large electronic contributions to the specific heat ($C/T \approx 1200$ mJ mol^{-1} K^{-2} below $T = 1.4$ K) and it is regarded as a heavy-fermion compound [2–5]. At higher temperatures ($T > 30$ K) the susceptibility is described by a Curie–Weiss law and the measured effective moment agrees well with the expectation of a Hund rule ground state for a free trivalent Ce ion [1, 2]. The susceptibility tends to a constant value $\chi(0) = 36 \times 10^{-3}$ emu mol^{-1} below 1.5 K [2] as is characteristic for many heavy-fermion systems. The initial report on the specific heat C of CeCuIn₂ [2] indicates a broad maximum near 2.5 K with no evidence of a magnetic transition. Subsequent studies on single-crystal CeInCu₂ [3] show that the specific heat exhibits a minimum at 8 K, increases rapidly with decreasing temperature and approximately saturates below 1 K, but with some fine structure of a peak at 0.8 K and another one at 0.17 K. It was also found that the magnetic susceptibility and Hall coefficient each exhibits a small peak at about 1 K [3]. The electrical resistivity, ρ, of CeInCu₂ increases upon cooling from room temperature, i.e. it exhibits a negative temperature coefficient of resistivity (TCR), goes through a peak at $T_{max} \sim 20$ K and then drops rapidly towards low temperatures [2]. Below 2.5 K $\rho(T)$ exhibits a T^2–Fermi liquid type of behaviour. The magnetic part of the resistivity (obtained after subtracting the resistivity of the non-magnetic LaInCu₂ homologue) shows a logarithmic Kondo-like increase upon cooling in the high-temperature regime ($T \geqslant 100$ K) [4].

   Although initial studies indicated no magnetic ordering in CeInCu₂, evidence of such order was first observed from NMR studies [5, 6] and corroborated by neutron diffraction measurements [7, 8]. Evidence is found from the ^{63}Cu NMR measurements of a random distribution between Ce and In atoms in the Heusler-type structure. Such a random distribution may be the source of the large residual resistivity observed for CeInCu₂ samples

[2–4]. Magnetic order which is of the type I structure of an fcc antiferromagnet occurs below $T_N = 2.3$ K. The neutron scattering is of a diffuse nature indicating very short-range order ($\xi_\parallel = 48$ Å, $\xi_\perp = 13$ Å) and with a moment $\mu = 0.40 \pm 0.07$ μ_B. A finite, but somewhat longer range of antiferromagnetic order has previously been observed in the heavy-fermion small-moment materials UPt$_3$ ($\xi \sim 250$ Å [9]) and URu$_2$Si$_2$ ($\xi \sim 150$ Å [10]).

Single-ion Kondo theory is often used as a departure point for describing heavy-fermion materials. The Kondo temperature T_K separates a high-temperature magnetic region ($T \gg T_K$) with $\ln T$ terms in the resistivity and specific heat from a low-temperature Fermi-liquid region ($T \ll T_K$) with power laws of the thermodynamic quantities in terms of temperature or magnetic field. T_K represents the only relevant energy scale for the system for all temperatures and magnetic fields. Notwithstanding this no self-consistent prescription exists for evaluating T_K from experimental data and somewhat different values of T_K are obtained from different experimental techniques and following various procedures [11]. Inelastic neutron scattering data have been used to find T_K for CeInCu$_2$. A Γ_7–Γ_8 crystal field splitting $\Delta = 90$ K has been observed for this compound together with a residual quasielastic linewidth $\Gamma_0 = 3.5$ K. The latter value leads to an estimated value of $T_K = \pi\Gamma_0/4$ [12] of 2.7 K. The measured susceptibility for $T < 30$ K deviates from a Curie–Weiss law and its temperature dependence may be explained by a crystal field splitting $\Delta = 98$ K, thus corroborating the neutron scattering result. From both susceptibility and specific results it has been concluded that the Γ_7 doublet is the ground state. Using $\Delta = 90$ K in combination with $\chi(0) = 36 \times 10^{-3}$ emu mol^{-1} has indicated a value $T_K = 6.5$ K [2]. Theoretical results for a dilute Kondo system allow estimates of T_K through the temperature of the specific heat maximum $T_K = 2.22T_{max}$ and from the $\gamma(T \to 0)$ value [13]. These give respectively $T_K = 5.6$ K and $T_K = 6.7$ K. Thus, T_K is observed as relatively small and of the same order of magnitude as the Néel temperature. Consequently one may expect to observe the effects of competition between the Rudermann–Kittel and Kondo interactions in this material.

In order to shed further light on the properties of CeInCu$_2$ a series of experiments on thermal expansion [14–19], elastic constants [17], electrical resistivity under pressure [14, 20, 21], and magnetoresistivity and magnetostriction under pressure [22] have been reported. Oomi *et al* [15] employing a strain gauge technique between 4 and 300 K found that the magnetic part of the thermal expansion coefficient of CeInCu$_2$, obtained after correcting for the phonon part, increases rapidly with decreasing temperature and goes through a peak at 25 K [16]. They interpreted this as originating from a Schottky anomaly as a result of the population of the first excited doublet crystal field state which they estimate at $\Delta = 63$ K. However, subsequent capacitance dilatometry studies of the thermal expansion coefficient α by Matsui *et al* [17] and de Visser *et al* [18] indicate a large broad magnetic contribution with a maximum that is centred at $T_{max} = 4.5$ K. Upon further decrease in temperature α becomes negative with a minimum centred at 1 K [18]. It has been suggested that the low-temperature anomaly in α is related to the short-range antiferromagnetic order of the material [18].

A 6% softening of the elastic bulk modulus C_B from 130 K down to 1.3 K has been deduced from ultrasonic measurements on single-crystal CeInCu$_2$ [17]. However, the authors observed no such softening of the transverse $(C_{11} - C_{12})/2$ and C_{44} modes. For dense Kondo 4f-electron compounds with relatively small values of T_K the elastic properties at temperatures above T_K usually exhibit the effect of quadrupolar interaction. Thus, softenings of $(C_{11} - C_{12})/2$ and C_{44} modes are observed for dense Kondo compounds such as CeAl$_2$ [23] and CeTe [24] which both have the Γ_7 ground state and these anomalies are explained by quadrupolar response theory [25]. The authors of [17] recognized that

compared to other dense Kondo systems with low T_K, CeInCu$_2$ behaves anomalously in that crystal electric field effects are not observed in its shear elastic constants.

The low-temperature peak in $\rho(T)$ for CeInCu$_2$ shifts to higher temperatures with application of pressure. The peak is furthermore smeared out at high pressures ($\geqslant$6 GPa) and $\rho(T)$ at high pressure is reminiscent of the behaviour of intermediate-valence materials [14, 20, 21]. Application of pressure also decreases the anomalous thermal expansion of CeInCu$_2$ at low temperatures [19].

Only a few alloying studies have been made on the CeInCu$_2$ system [2, 4]. Najib *et al* [4] substituted La and Y on Ce sites and found two terms in the resistivity, viz. a one-impurity and a two-impurity contribution. The latter term describes the intersite correlations between Ce atoms and becomes important for Ce concentrated systems. Ligand substitutions were made only on Cu sites and with the isoelectronic Ag element [2]. Such a substitution leaves the Ce sublattice unperturbed and should only influence the atomic volume. Indeed, alloying with Ag leads to an increase in unit-cell volume and a decrease in the magnitude of the coupling constant between the 4f state and the conduction band. As a consequence, the Néel temperature, T_N, was observed to increase in agreement with Doniach's Kondo lattice model [26].

In the present work we study the effect of alloying on the In sites by M = Al, Ga and Sn substitutions and on the Cu sites by T = Fe, Ni, Pd and Pt substitutions. The change in the room-temperature lattice parameter and the temperature dependence of the electrical resistivity were investigated. In the case of Al or Ga substitutions, the electron concentration, n_e, in the alloys remains constant, suggesting that the resulting change in the physical properties is expected due to a change in the cell volume, ΔV. The remaining doped alloys could behave in a more complex manner, depending on the magnitude of both ΔV and the change in electron concentration, Δn_e.

2. Experiment

The alloys CeIn$_{1-x}$M$_x$Cu$_2$ and CeIn(Cu$_{1-x}$T$_x$)$_2$ with different values of x in the range 0.1–0.2, were prepared by arc-melting stoichiometric amounts of the constituent elements in purified argon atmosphere. The wt% purities of the materials were 6N, except for Ce which was 4N. The resulting ingots were turned over and re-melted several times to obtain homogeneous samples. The specimens were further examined by powder x-ray diffraction at room temperature using Cu Kα radiation. Samples for the resistivity measurements were cut from the ingots using spark erosion and had typical dimensions of $1.0 \times 1.0 \times 10$ mm^3.

Electrical resistivity, ρ, was measured by a standard four-probe method. A constant DC current of 100 mA was passed through the sample and a voltmeter of 0.1 μV resolution was used to measure the voltage drop over the sample. The absolute accuracy of determining ρ was within 5%, the main uncertainty being in the geometrical factors. The data were collected in the temperature range 4.2–300 K during cooling as well as heating runs and temperatures were measured using an Au–0.07 at.% Fe versus chromel thermocouple. No correction was made for the effect of thermal expansion (about 0.5% over the temperature range 4–300 K) on the calculated values of $\rho(T)$.

3. Results and discussion

For CeInCu$_2$ x-ray diffraction patterns have been observed which are characteristic of the $L2_1$ face centred cubic type structure and show no evidence of any secondary phase. The

lattice parameter of our sample is $a = 6.789$ (± 0.003) Å, and is consistent with the data reported by Felner [1], Lahiouel *et al* [2] and Takagi *et al* [5]. For solid solutions $CeIn_{1-x}M_xCu_2$ and $CeIn(Cu_{1-x}T_x)_2$ the x-ray diffraction studies also showed reflections belonging to the cubic structure of $CeInCu_2$. However, the diffraction patterns of some alloys with T = Ni, Pd and Pt indicate weak additional reflections. The amount of unknown impurity phases, estimated from the reflection intensities is about 5–10%.
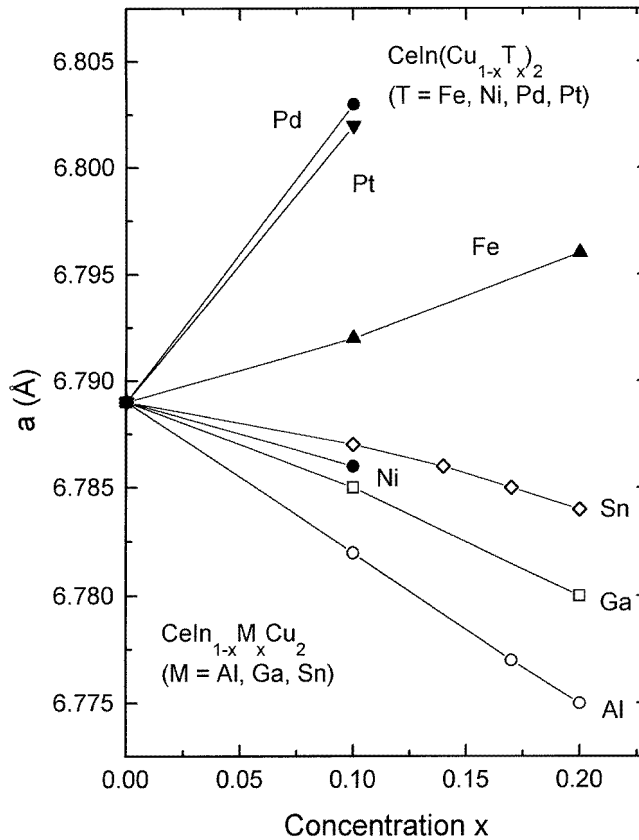

Figure 1. The lattice parameters of $CeIn_{1-x}M_xCu_2$ and $CeIn(Cu_{1-x}T_x)_2$ compounds as a function of x for M = Al, Ga and Sn, and T = Fe, Ni, Pd and Pt.

The variations of the lattice parameter with concentration, $a(x)$, for the solid solutions $CeIn_{1-x}M_xCu_2$ and $CeIn(Cu_{1-x}T_x)_2$ are plotted in figure 1. Substitution of metalloid atoms, Al, Ga or Sn, decrease the lattice parameter, resulting in chemical compression in these alloys. The largest change is observed for Al doped alloys, since Al has the smallest atomic radius in comparison with that of Ga or Sn. Substitutions by transition metal atoms lead to a small decrease in the lattice parameter for Ni doping, while for Fe, Pd or Pt doping the lattice parameter is increased.

In figure 2 our measured $\rho(T)$ for $CeInCu_2$ is displayed and it confirms the existence of a negative TCR at high temperatures and a low-temperature maximum [2, 3]. The maximum in $\rho(T)$ is observed at $T_{max} = 19$ K in our sample compared to $T_{max} \approx 23$ K in previous studies [2, 3]. Our room-temperature resistivity, $\rho(RT)$, of 92 $\mu\Omega$ cm is comparable with 100 $\mu\Omega$ cm measured by Onuki *et al* [3] but smaller than 135 $\mu\Omega$ cm measured by Lahiouel
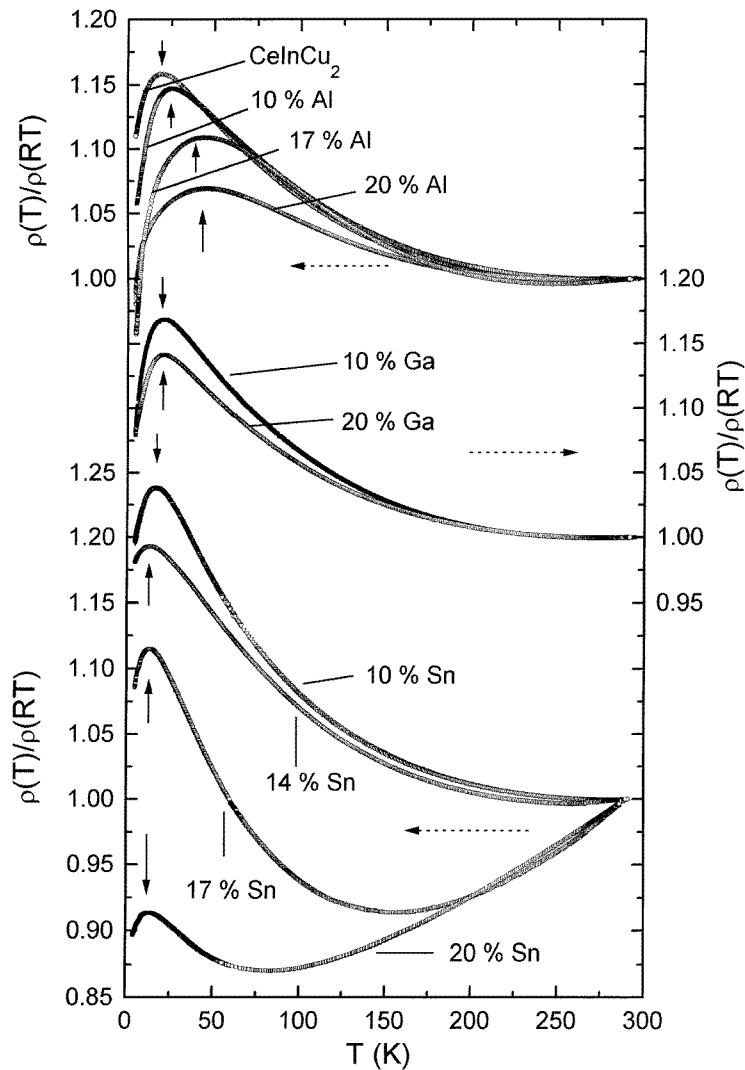
Figure 2. The temperature dependences of the electrical resistivity normalized to the values at room temperature for $CeIn_{1-x}M_xCu_2$ (M = Al, Ga and Sn). The arrows show the position of the resistivity maximum, T_{max}.

et al [2]. The ratio $\rho(T_{max})/\rho(RT) = 1.16$ in our case is comparable with that measured on a single-crystal sample by Onuki *et al* [3].

Figure 2 also shows the temperature dependence of the electrical resistivity for the $CeIn_{1-x}Al_xCu_2$, $CeIn_{1-x}Ga_xCu_2$ and $CeIn_{1-x}Sn_xCu_2$ alloys. The substitution of In with Al strongly affects the resistivity behaviour of $CeIn_{1-x}Al_xCu_2$. It is observed that the substitutions suppress the slope $d\rho/dT$ at high temperatures and the $\rho(T_{max})/\rho(RT)$ ratio, and enlarge the room-temperature resistivity (table 1). The latter effect may be explained by an increase of the atomic disorder scattering due to Al substitutions.

The primary effect of In substitution by Al in $CeIn_{1-x}Al_xCu_2$, however, is that the system shows an increase in the value of the temperature T_{max}. Two possible mechanisms

Table 1. Transport characteristics of $CeIn_{1-x}M_xCu_2$ and $CeIn(Cu_{1-x}T_x)_2$ intermetallics (M = Al, Ga and Sn, and T = Fe, Ni, Pd and Pt).

Compounds	T_{max} (K)	$\rho(T_{max})/\rho(RT)$	$\rho(RT)$ ($\mu\Omega$ cm)
$CeInCu_2$	19.0	1.16	92
$CeIn_{0.90}Al_{0.10}Cu_2$	24.9	1.15	95
$CeIn_{0.83}Al_{0.17}Cu_2$	38.7	1.11	98
$CeIn_{0.80}Al_{0.20}Cu_2$	42.0	1.07	121
$CeIn_{0.90}Ga_{0.10}Cu_2$	19.2	1.16	100
$CeIn_{0.80}Ga_{0.20}Cu_2$	19.5	1.14	139
$CeIn_{0.90}Sn_{0.10}Cu_2$	16.0	1.24	97
$CeIn_{0.86}Sn_{0.14}Cu_2$	13.5	1.19	122
$CeIn_{0.83}Sn_{0.17}Cu_2$	12.8	1.13	137
$CeIn_{0.80}Sn_{0.20}Cu_2$	11.6	0.92	352
$CeIn(Cu_{0.90}Fe_{0.10})_2$	20.3	1.14	100
$CeIn(Cu_{0.80}Fe_{0.20})_2$	24.2	1.03	109
$CeIn(Cu_{0.90}Ni_{0.10})_2$	21.2	1.14	120
$CeIn(CuO_{0.90}Pd_{0.10})_2$	22.9	1.55	115
$CeIn(Cu_{0.90}Pt_{0.10})_2$	33.7	1.14	107

may be responsible for this effect, viz. the enhancement of the Kondo temperature and/or the increase in the crystal field splitting. These two mechanisms will be discussed in the following.

Lavagna *et al* [27] have shown that the temperature T_{max} of Kondo-lattice systems is closely related to T_K. Hence, it follows that

$$T_{max} \propto T_K \propto \exp(-1/|JN(E_F)|) \tag{1}$$

where $N(E_F)$ is the density of states at the Fermi level and the Schrieffer–Wolff transformation of the Anderson model gives the exchange integral as

$$J = \frac{\langle V_{kf}^2 \rangle U}{\varepsilon_f(\varepsilon_f + U)}. \tag{2}$$

In (2) V_{kf} is the matrix element characterizing the hybridization between 4f and conduction electrons at E_F, U is the Coulomb integral and ε_f is the position of the 4f level relative to the Fermi level. In our $CeIn_{1-x}Al_xCu_2$ system, the substitution decreases the volume of the unit cell. This can lead the system to increase $|JN(E_F)|$ and thus enlarge T_K.

In the compressible Kondo model [29] $|JN(E_F)|$ is assumed to have the following volume dependence:

$$|JN(E_F)| = |JN(E_F)|_0 \exp(-q(V - V_0)/V_0). \tag{3}$$

In (3) $|JN(E_F)|_0$ indicates the value of the quantity at initial volume V_0 and $|JN(E_F)|$ corresponds to volume V. The numerical constant q takes a value between 6 and 8. Expanding the exponential in (3) for small values of $\Delta V = V - V_0$ and substituting the result in (1) leads to

$$\ln[T_{max}(V)/T_{max}(V_0)] = \frac{-q\Delta V}{V_0|JN(E_F)|_0}. \tag{4}$$

In figure 3 a plot of the left-hand side of (4) against $(-\Delta V/V_0)$ is given where our x-ray measurements have been used to obtain the latter quantity. A satisfactory fit to the equation is indicated. Using the slope from figure 3 and $q = 6$ gives $|JN(E_F)|_0 = 0.047$. This
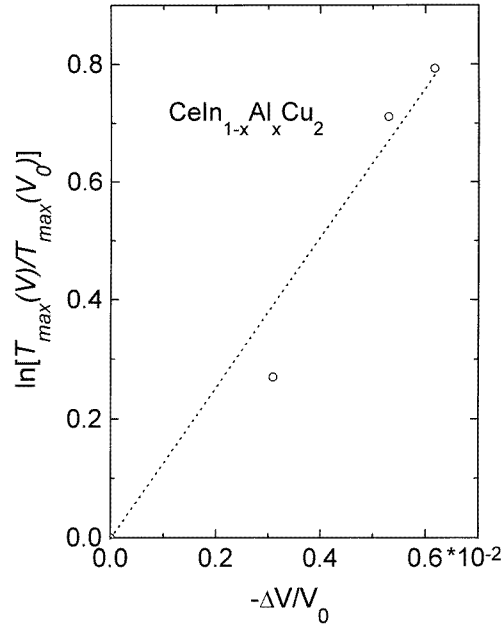
Figure 3. Plot of $\ln[T_{max}(V)/T_{max}(V_0)]$ against $(-\Delta V/V_0)$ in order to ascertain the validity of (4) and giving a value of $|JN(E_F)|_0 = 0.047$ (see text).

value is somewhat lower than $|JN(E_F)|_0 = 0.081$ obtained by Kagayama *et al* [20] for CeInCu₂ from resistivity measurements under hydrostatic pressure.

We want to discuss another aspect of Al substitution, namely the possible influence of volume change on the crystalline electric field (CEF) effects. The importance of the CEF to the electrical resistivity of Ce compounds that exhibit the Kondo effect had already been indicated early on by Maranzana and Bianchessi [30, 31] and a comprehensive theoretical model was developed by Cornut and Coqblin [32]. By considering Ce ions with $J = 5/2$ for which a hexagonal crystal field splits the energy levels of the spin into three doublets $\pm 1/2$, $\pm 3/2$ and $\pm 5/2$ and with the $\pm 5/2$ doublet as ground state, Maranzana showed that no Kondo resonances occur at the Fermi level, but rather at the CEF energy splitting $\pm \Delta_{21}$ between 5/2 and 3/2 (or $-5/2$ and $-3/2$) states. The Kondo effect is then expected to appear at temperatures larger than Δ_{21}/k_B. Cornut and Coqblin formulated the Anderson Hamiltonian with inclusion of the CEF effect appropriate for Ce impurities, derived the Schrieffer–Wolff transformation for this Hamiltonian and calculated the Kondo resistivity for such systems. The prediction of theory is that if there exist a number of CEF levels Δ_{21}, Δ_{31}, ... well separated from each other and from the ground state, then the resistivity will go through a broad peak around each Δ_{i1} value, exhibit a $\ln T$ behaviour at high temperatures, possibly an intermediate $\ln T$ behaviour if Δ_{31} and Δ_{21} are very well separated and always a $\ln T$ behaviour at low temperatures. The ratio of low-temperature to high-temperature slopes is predicted as 3/35 if the ground state of the Ce impurity is a doublet and 3/7 if the ground state is a quartet. The behaviour at high temperatures, which is associated with the CEF splittings, will be described by the high-temperature Kondo temperature T_K^H. Some of the features predicted by Cornut and Coqblin are at least qualitatively observed for certain Ce compounds, e.g. for CeCu₂Si₂ [33], which behaves as $\ln T$ at high temperatures, goes through a maximum at 100 K (associated with the CEF overall splitting [34]) and with

further decrease in temperature goes through a second broad maximum at 20 K, this being associated with the low-temperature Kondo behaviour [34]. The resistivity of CeAl$_3$ [35] goes through a single peak at $T_{max} \approx 30$ K (associated with crystal field effects) and then continuously decreases with decreasing temperature to the lowest measured temperatures. Employing a phenomenological form for the CEF splitting Lassailly *et al* [34] simulated the form of the observed $\rho(T)$ for CeAl$_3$ through a choice of parameters that include $\Delta = 100$ K and $T_K = 5$ K.

No attempt was made for our CeInCu$_2$ system to model the observed $\rho(T)$ in terms of the above theory [32, 34], since $\rho(T)$ measurements have only been performed to 4 K and the system would in any case exhibit complications of magnetic order and high values of residual resistivity for $T < 4$ K. Rather consider the relationship $T_K^H = (T_K \Delta_{12} \Delta_{13})^{1/3}$ derived by Hanzawa *et al* [36], in which Δ_{12} and Δ_{13} denote the CEF splitting between the ground state and the first and second excited levels, respectively. To derive an order of magnitude estimate of T_K, we use the value of $T_{max} = 19$ K observed for CeInCu$_2$ for T_K^H in the preceding equation, take $\Delta_{12} = \Delta_{13} = \Delta = 90$ K (from neutron scattering) and hence find $T_K = 0.85$ K. If on the other hand $\Delta = 63$ K [15] is used a value of $T_K = 1.7$ K is obtained. These values agree in order of magnitude with $T_K = 2.7$ K obtained from neutron inelastic scattering. It has been observed for some Ce based compounds that the effect of the CEF on the resistivity behaviour can be altered by applied pressure [37, 38]. It would therefore seem reasonable to argue that the shift in T_{max} to high temperatures with increasing Al-concentration may indicate an increase in CEF splitting. However, the relative contributions to the change in T_{max} that should be apportioned to the volume dependences of T_K or of the CEF splitting are difficult to determine.

The influence of the other substitutions will now be discussed. In the case of CeIn$_{1-x}$Ga$_x$Cu$_2$, the effect of Ga substitution on the $\rho(T)$ behaviour of CeInCu$_2$ is less pronounced than that for Al substitution (figure 2). For all the Ga based alloys the single-impurity Kondo behaviour at high temperatures and the maximum of resistivity at lower temperatures persist and T_{max} is increased only to about 19.5 K at $x = 0.2$. This apparent invariance of the magnetic and CEF interactions in spite of a considerable change in volume with Ga substitution is surprising and an explanation for this behaviour will have to await further studies.

The behaviour of CeIn$_{1-x}$Sn$_x$Cu$_2$ is quite different from the Al or Ga doped alloys, namely when In is substituted by Sn the position of T_{max} is shifted towards lower temperatures. Furthermore, the minimum characteristic of the Kondo effect is shifted to low temperatures, indicating a weakening of the Kondo interaction. This gives rise to a change in sign of the TCR for the $x = 0.17$ and $x = 0.20$ alloys above 160 and 80 K, respectively. The observed behaviour cannot be attributed to a reduction of the unit-cell volume, since this is decreased with increasing x and should give rise to an increase of V_{kf}. Of greater consequence rather is the effect of the increase in the number of conduction electrons due to Sn substitution. Possibly, the increase Δn_e raises the Fermi level E_F and increases the magnitude of ε_f. Thus, in the Sn based alloys there could be a competition between the effects caused by the increase of ε_f and V_{kf}, and if the first one is the major effect, the final result will be a decrease of T_K and Δ and hence of T_{max}.

Complementary information about the influence of substitution on the physical properties of CeInCu$_2$ can be obtained from the CeIn(Cu$_{1-x}$T$_x$)$_2$ alloys, where alloying on the Cu sites can modify the d band and, therefore, the strength of hybridization between the 4f and d states. When Cu atoms are replaced by the transition metal atoms, the number of d electrons decreases. This may lead the system to be more delocalized and destroy the magnetic ordering in CeInCu$_2$. Further experiments are planned to support this suggestion.

On the other hand, the decrease in the number of d electrons at the Fermi level is expected to decrease the magnitude of the energy ε_f and thus to increase the temperature T_K. As is evident from figure 4 and table 1 an increase in T_{max} is found for all $CeIn(Cu_{1-x}T_x)_2$ substitutions studied here with the most pronounced effect observed for the Pt substitution.
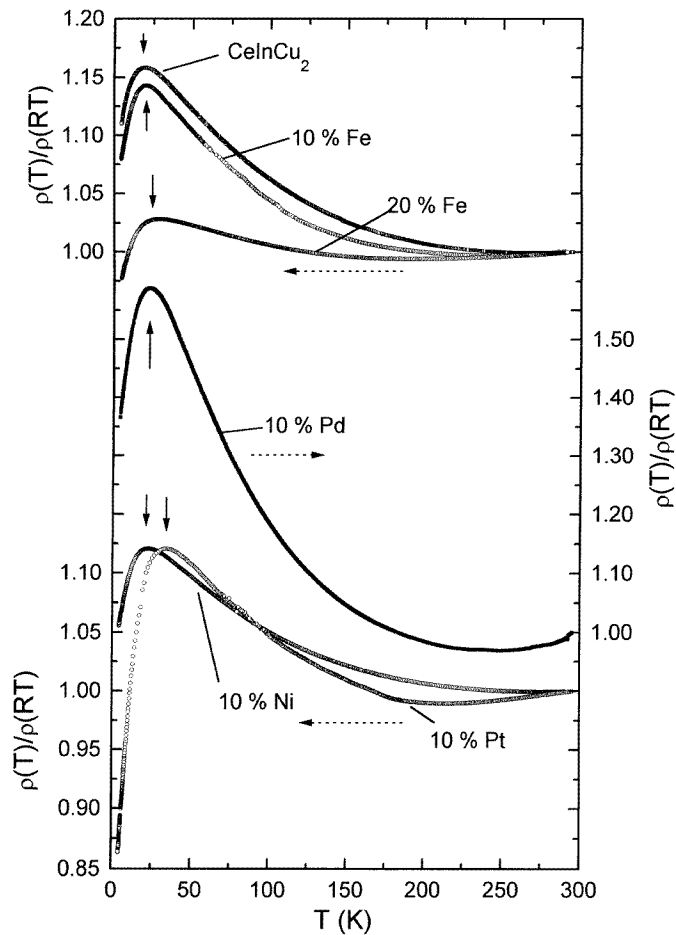

Figure 4. The temperature dependences of the electrical resistivity normalized to the values at room temperature for $CeIn(Cu_{1-x}T_x)_2$ (T = Fe, Ni, Pd and Pt). The arrows show the position of the resistivity maximum, T_{max}.

The substitution of Cu by a transition metal in $CeInCu_2$ may also cause volume related effects. The larger volume of alloys containing Fe, Pd or Pt compared to $CeInCu_2$ may lead these systems to have smaller values of V_{kf} tending to decrease T_{max}. However, similar to the case of Sn doping these volume effects are not expected to dominate.

Finally, it is noted that whereas for most substitutions $\rho(T_{max})$ is about 10 or 15% higher than at room temperature, it shows a 55% increase for the $CeIn(CuO_{0.90}Pd_{0.10})_2$ alloy. This aspect warrants further investigation.

4. Conclusions

We have studied the effects of Al, Ga, Sn or Fe, Ni, Pd and Pt doping in the cubic $CeInCu_2$ compound. At least 20% of In or 10% of Cu can be replaced by respective metalloid or transition metal atoms without changing the crystal structure. The main influence of substitutions on the physical properties of $CeInCu_2$ is to change the Kondo temperature and possibly also the CEF energy splitting. These values depend on the number of conduction electrons in the alloys and on the volume of the unit cell.

Acknowledgments

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