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SUBSTITUTION STUDY ON THE COHERENT KONDO STATE IN $CeCu_5In$

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Ternary alloys $CeCu_{5-x}In_{1+x}$ were studied by means of magnetic and electrical resistivity measurements. In $CeCu_5In$ a coherent Kondo state without long-range magnetic order develops at low temperatures. It transforms into an incoherent Kondo state upon small substitution of In for Cu.

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1. Introduction

The compound $CeCu_6$ is an archetypal non-magnetic heavy fermion system [1]. The discovery of a non-Fermi liquid behavior in the series $CeCu_{6-x}Au_x$ [2] has focused attention on other solid solutions of this type. Recently, Kasaya et al. [3] reported on the alloys $CeCu_{6-x}In_x$ with $0 \leq x \leq 1.75$, claiming that they are isostructural to $CeCu_6$. Contrary to that, we showed before [4] that $CeCu_{4.38}In_{1.62}$ crystallizes with its own structure type, which is a ternary derivative of the $CeCu_6$-type. In all the alloys $CeCu_{6-x}In_x$ but $CeCu_5In$ two crystallographic sites have mixed Cu/In occupancy. In the latter compound only there occurs a full atom order and owing to that a coherent Kondo behavior develops [3]. In this paper we report on the destruction of coherent state upon Cu → In exchange.

2. Experimental details

Polycrystalline samples of $CeCu_{5-x}In_{1+x}$ alloys with $x = 0.0, 0.2, 0.4$, and 0.6 were prepared by arc melting the appropriate amounts of constituting elements (nominal purity: Ce — 99.85 mass%, Cu — 99.92 mass%, In — 99.99 mass%) under a titanium gettered argon atmosphere and subsequent annealing in vacuum at 870 K for 20 days. X-ray powder diffraction analyses revealed all the alloys to be single phase with the $CeCu_{4.38}In_{1.62}$-type crystal structure. The orthorhombic lattice parameters increase with rising x, in agreement with Ref. [3] (see Table).

Magnetic measurements were carried out in the temperature range 1.7–300 K and in magnetic fields up to 5 T employing a Quantum Design SQUID magnetometer. The electrical resistivity was measured from 4.2 K to 300 K using a conventional dc four-point technique.

TABLE

Lattice parameters and magnetic data for $CeCu_{5-x}In_{1+x}$ alloys.

x	a [nm]	b [nm]	c [nm]	μ_{eff} [μ_B]	$-\theta_p$ [K]	μ_{5T} [μ_B]
0.0	1.6720(4)	1.0600(3)	0.5074(1)	2.68(9)	48(6)	0.13(3)
0.2	1.6767(3)	1.0674(3)	0.5101(1)	2.67(5)	45(4)	0.25(2)
0.4	1.6852(5)	1.0752(4)	0.5124(1)	2.64(6)	42(8)	0.53(2)
0.6	1.6921(4)	1.0801(3)	0.5159(2)	2.62(6)	22(7)	0.58(2)

3. Results and discussion

The inverse magnetic susceptibility of $CeCu_{5-x}In_{1+x}$ intermetallics is presented in Fig. 1. Above 40 K the susceptibility of all these alloys but $CeCu_{4.4}In_{1.6}$ has almost the same magnitude and exhibits (up to 300 K) a Curie–Weiss behavior with the parameters given in Table. The effective magnetic moments are close to that expected for a free Ce^{3+} ion ($2.54\mu_B$), and the paramagnetic Curie temperatures are negative and rather large. The susceptibility of $CeCu_{4.4}In_{1.6}$ also follows a Curie–Weiss law (above 50 K), with a similar value of μ_{eff} but with θ_p being only half the value found for the other phases. At the lowest temperatures, $\chi^{-1}(T)$ of all the alloys markedly deviates from a straight line, probably due to thermal depopulation of crystal field (CF) levels. The Cu $\rightarrow$ In exchange is accompanied by a change in the shape of low-temperature $\chi^{-1}(T)$, which reflects distinct modifications in the CF potential acting on the $Ce^2F_{\frac{5}{2}}$ ground multiplet.

For all the alloys studied no signature of any magnetic order is observed in $\chi(T)$ down to 1.7 K. This is in contrast to the specific heat results [3], which revealed antiferromagnetism in $CeCu_{4.5}In_{1.5}$ below 2.2 K. Yet, with rising x the magnetization measured at 1.7 K in a field of 5 T considerably increases, and $\sigma(T)$ of $CeCu_{4.4}In_{1.6}$ is strongly curvilinear (see the inset to Fig. 1 and Table).

In agreement with the previous finding [3], the electrical resistivity of $CeCu_5In$ reveals features characteristic of coherent Kondo effect (see Fig. 2). The broad maximum in $\rho(T)$, occurring at about 25 K, is consistent with $T_K \approx 28$ K, as derived from the specific heat data [3], and with $|\theta_p| = 48$ K, found in the magnetic measurements. In order to estimate the magnetic contribution to the resistivity of $CeCu_5In$ it was assumed that the phonon contribution $\rho_{ph}(T)$ can be approximated by the temperature-dependent resistivity of its isostructural lanthanum counterpart. As shown in Fig. 2, $\rho(T)$ of $LaCu_5In$ can be well described by the Bloch–Grüneisen–Mott (BGM) expression

$$\rho(T) = \rho_0 + \rho_{ph}(T) = \rho_0 + 4RT\left(\frac{T}{\Theta_D}\right)^4 \int_0^{\Theta_D/T} \frac{x^5 dx}{(e^x-1)(1-e^{-x})} - KT^3,$$

where ρ_0 is the residual resistivity, Θ_D stands for the Debye temperature and R is a constant, whereas the cubic term KT^3 describes interband scattering processes [5]. The least-squares fit parameters for $LaCu_5In$ are as follows: $\rho_0 = 8$ $\mu\Omega$ cm, $R = 0.14$ $\mu\Omega$ cm/K, $\Theta_D = 157$ K and $R = 2.9 \times 10^{-7}$ $\mu\Omega$ cm/K^3. The resulting temperature dependence of the magnetic resistivity of $CeCu_5In$ (enlarged by ρ_0)

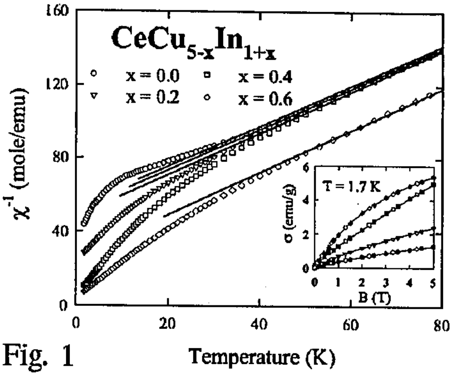

Fig. 1

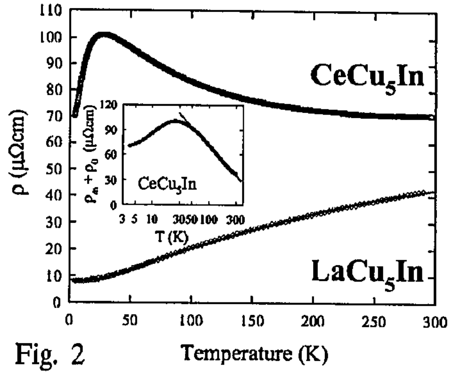

Fig. 2

Fig. 1. Low-temperature dependence of the inverse magnetic susceptibility of $CeCu_{5-x}In_{1+x}$ alloys. The solid lines are fits of $\chi^{-1}(T)$ to the Curie–Weiss law. The inset shows the field variation of the magnetization of $CeCu_{5-x}In_{1+x}$ alloys measured at 1.7 K with increasing and decreasing magnetic field (closed and open symbols, respectively).

Fig. 2. Temperature dependence of the electrical resistivity of $CeCu_5In$ and $LaCu_5In$. The solid curve is a fit of $\rho(T)$ of $LaCu_5In$ to the BGM expression. The inset shows the magnetic contribution to the resistivity of $CeCu_5In$. The solid line marks here a $-\ln T$ variation of ρ_m.

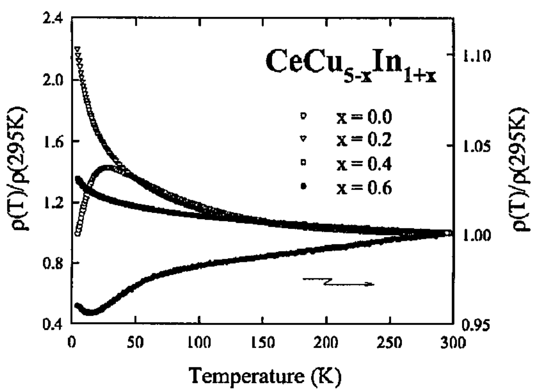

Fig. 3. Temperature variation of the normalised electrical resistivity of $CeCu_{5-x}In_{1+x}$ alloys. Note different ordinate scale for $x = 0.6$.

is shown in the inset to Fig. 2. Above 50 K it follows the Kondo formula

$$\rho_m(T) = \rho_0^\infty - c_K \ln T$$

with a value of 220 $\mu\Omega$ cm for the sum $\rho_0 + \rho_0^\infty$ (ρ_0^∞ is the spin-disorder resistivity) and the Kondo coefficient $c_K = 32$ $\mu\Omega$ cm.

Pronounced Kondo effect dominates also the electrical properties of the $CeCu_{5-x}In_{1+x}$ alloys with $x = 0.2$ and 0.4 (see Fig. 3). Both these intermetallics exhibit a negative temperature coefficient in $\rho(T)$ in the whole temperature range studied. A similar procedure of extracting $\rho_m(T)$, applied using the resistivity data of the respective $LaCu_{5-x}In_{1+x}$ alloys, gave the Kondo coefficients $c_K = 49$ and

36 $\mu\Omega$ cm for $x = 0.2$ and 0.4 respectively. The resistivity of $CeCu_{4.4}In_{1.6}$ is hardly temperature dependent. The Kondo contribution with $c_K = 13$ $\mu\Omega$ cm is here too small to overcome the phonon term and therefore a negative $\partial\rho/\partial T$ is seen at the lowest temperatures only. It is worthwhile noting that apparent weakening of the Kondo interactions along the $CeCu_{5-x}In_{1+x}$ series is reflected also in the values of θ_p, which become less negative with rising x (see Table).

The most spectacular feature, observed in Fig. 3, is a rapid destruction of the coherent Kondo state in $CeCu_5In$ upon even small substitution of In for Cu. Somewhat different behavior was recently established for the magnetically diluted alloys $Ce_{1-x}La_xCu_5In$ as well as for the alloys $CeCu_5In_{1-x}Ga_x$ and $CeCu_5In_{1-x}Al_x$ [6]. Characteristic of all these latter series is a gradual evolution from coherent Kondo to incoherent Kondo state with increasing x. Apparently, the Kondo behavior in $CeCu_5In$ is less sensitive to atom disorder in the Ce and In sublattices than in the Cu sublattice. This finding may suggest that the magnetic properties of the alloys investigated are governed mainly by the f–d hybridization.

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