Characterization of the mixed-valence to heavy-fermion transition in $CeCu_{2-x}Co_xSi_2$ by susceptibility and resistance studies

S. K. Dhar, E. V. Sampathkumaran, J. Ray, and G. Chandra
Tata Institute of Fundamental Research, Colaba, Bombay 400005, India
(Received 22 December 1986)

The transformation of the heavy-fermion ground state to the mixed-valence state is studied in the pseudoternary alloys $CeCu_{2-x}Co_xSi_2$ by lattice-parameter, susceptibility, and resistivity measurements. The results support our earlier conclusions on the relationship between the lattice parameter, low-temperature susceptibility, resistivity, and Kondo temperature in Kondo-lattice systems and are qualitatively consistent with the recent theoretical predictions of Auerbach and Levin.

One of the important aspects of rare-earth and actinide research is to understand the factors controlling the formation of the heavy-fermion ground state.[1,2] In this respect, solid-solution studies involving the replacement of Cu by Ni in $CeCu_2Si_2$ have yielded interesting results.[3,4] It was noticed[3] that the substitution of about 30% of Cu by Ni resulted in a drastic depression in the low-temperature susceptibility $\chi(T\rightarrow 0)$ and the electronic specific heat γ.[5] The Ce (non-$4f$)-ligand distance, which is determined by the c parameter of the unit cell, also undergoes a faster decrease[3] in the same interval, tracking the x dependence of $\chi(0)$ and γ. The double-peaked structure in the resistivity smears out with the addition of nickel. The observed correlations in the physical properties are quite helpful in understanding the heavy-fermion ground state. In order to put our earlier conclusions on a firm footing, we have investigated another pseudoternary series $CeCu_{2-x}Co_xSi_2$, by lattice-parameter, susceptibility (χ), and resistivity (ρ) measurements and the results are in conformity with earlier conclusions.[3,4]

Polycrystalline samples of $CeCu_{2-x}Co_xSi_2$ alloys ($x=0$, 0.15, 0.3, 0.5, 0.8, and 2.0) were prepared by arc melting together the stoichiometric amounts of the constituent elements and were characterized by x-ray diffraction. Magnetic susceptibility (χ) measurements were performed in the temperature interval 4.2–300 K in a field of 6 kOe employing a Faraday balance. A standard four-probe technique using a Keithley dc nanovoltmeter was employed down to 0.5 K to measure the electrical resistivity. Other experimental details of this measurement may be obtained from Ref. 4.

The results of the lattice parameter, magnetic susceptibility (χ), and resistance (ρ) measurements are presented in Figs. 1–3. The observed features are very much similar to those reported for nickel-substituted alloys.[3,4] The lattice parameter a is a linear function of x, whereas c undergoes a faster reduction for $x \le 0.5$. The susceptibility at 4.2 K, a measure of $\chi(0)$, decreases rapidly with the replacement of Cu by Co and for $x \gtrsim 0.5$, χ is nearly temperature independent, typical of strongly mixed-valence materials. The x dependence of $\chi(0)$, a measure of heavy-fermion behavior, tracks that of the lattice parameter c (related to Ce-ligand distances). This correlation supports the earlier idea[3] that the heavy-fermion ground state is less hybridized than the mixed-valence ground state. Besides, the continuous evolution of the heavy-fermion ground state in $CeCu_2Si_2$ from the strongly mixed-valence state in $CeCo_2Si_2$ implies that a single mechanism controls the low-temperature properties of all these phenomena.[6]

The resistive behavior (Fig. 3) is also consistent with a transformation of the heavy-fermion state to the mixed valence state by the substitution of Co for Cu in $CeCu_2Si_2$. The double-peaked structure seen in $CeCu_2Si_2$ merges into a single peak as x is increased. For larger values of x, we find that the peak due to crystal fields[7,8] (at about 100 K) disappears, and we infer from this observation that crystal fields do not survive under strong-valence fluctuations. An important point to be made is that the position of the resistance peak on the low-temperature side (20 K in $CeCu_2Si_2$) moves so fast with x to higher temperatures that it is not resolvable from that due to crystal fields (even for $x=0.15$); this peak posi-

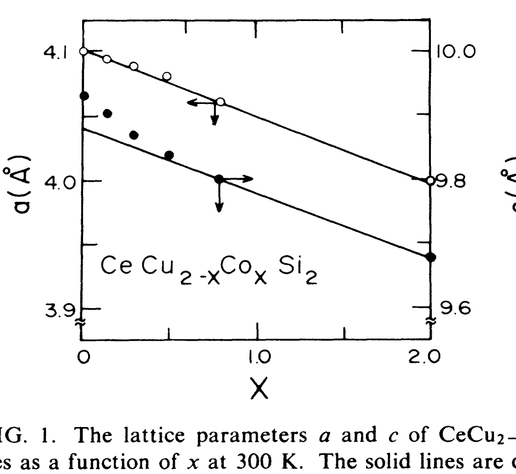

FIG. 1. The lattice parameters a and c of $CeCu_{2-x}Co_xSi_2$ series as a function of x at 300 K. The solid lines are drawn to show the anomaly in c as $x\rightarrow 0$.

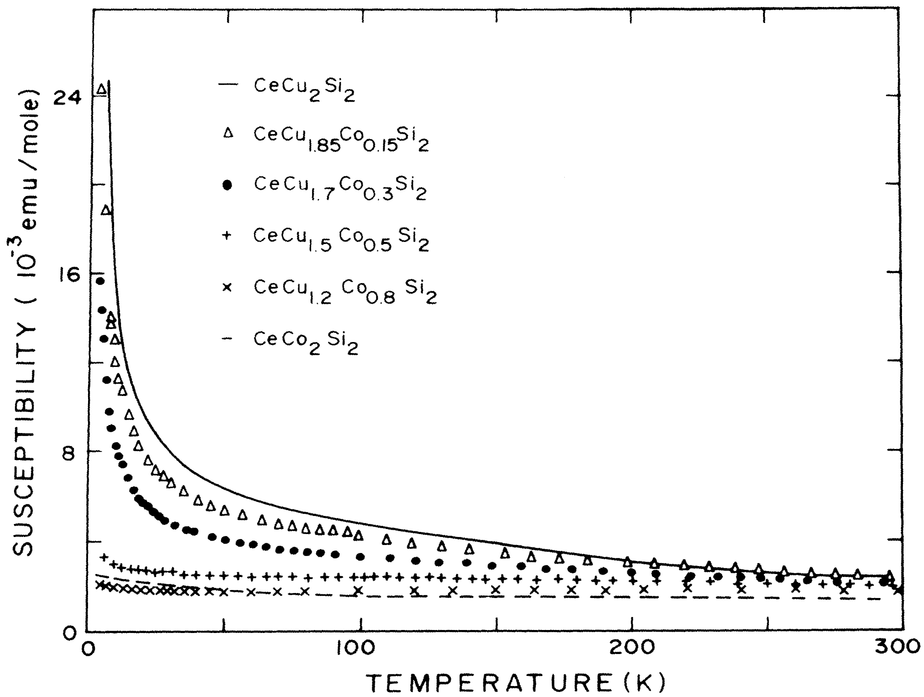

FIG. 2. Magnetic susceptibility as a function of temperature (4.2–300 K) for the alloys of the series $CeCu_{x-2}Co_xSi_2$.

tion when resolvable, as in the earlier system $CeCu_{2-x}Ni_xSi_2$,[4] tracks qualitatively the x dependence of $C/\chi(0)$ (here C is the Curie constant), a measure of Kondo temperature T_K. This observation is consistent with the traditional belief[9–11] that the low-temperature-size peak is related to T_K. We also notice that if one plots ρ vs T^2, the coefficient (A) of T^2 at low temperatures keeps decreasing with x with a drastic variation around $x=0.5$. By analyzing the ρ-T^2 data below 10 K, it is found that $A=9.1$ (in units of $\mu\Omega\,\mathrm{m/K^2}$) for $x=0$ dropping to $A=0.18$ for $x=2$.

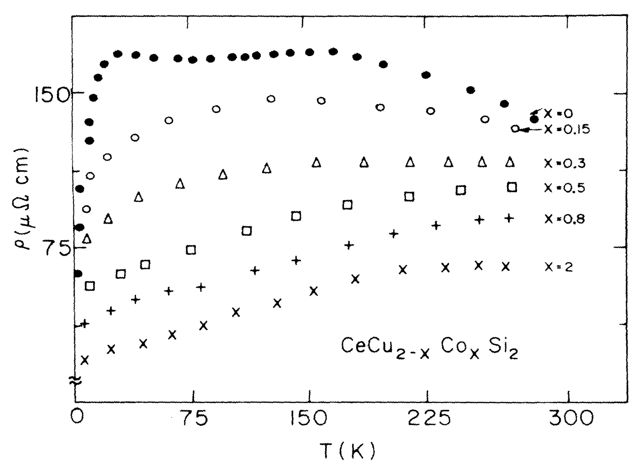

FIG. 3. Resistivity (ρ) as a function of temperature (0.5–300 K) for the alloys of the series $CeCu_{2-x}Co_xSi_2$.

From the results obtained for $CeCu_{2-x}T_xSi_2$ ($T=$Co and Ni), we believe that (i) the $4f$-hybridization strength for the heavy-fermion ground state is relatively smaller than the mixed valence ground state and a single mechanism controls both phenomena, (ii) crystal fields do not survive under the influence of strong-valence fluctuations, (iii) the position of the low-temperature peak in the resistivity of $CeCu_2Si_2$ is related to T_K contrary to the arguments of Schilling,[12] and (iv) there is a scaling relation between γ, χ, and A.[13] Further detailed arguments were presented elsewhere.[3,4] Our results are qualitatively consistent with the recent theoretical predictions of Auerbach and Levin[14] in that the theories involving multiple energy scales such as the Rudermann-Kittel-Kasuya-Yosida interaction temperature are incompatible with the simple scaling relations between γ, χ, and A.

Finally, we would like to remark that though the two related phenomena—heavy fermion and mixed valence—depend on Ce-ligand distances, the value of the critical distance could in principle vary from one compound to another.

We thank Professor R. Vijayaraghavan for his keen interest and encouragement in this work.

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