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Anomalous negative thermal expansion of $CeInCu_2$

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The coefficients of linear thermal expansion, $\alpha(T)$, of the heavy-fermion compound $CeInCu_2$ and its non-heavy analog $LaInCu_2$ have been measured at low temperatures ($0.3\,K < T < 12\,K$). In the heavy-fermion regime $\alpha(T)$ of $CeInCu_2$ shows a large positive contribution (with an electronic Grüneisen parameter $\Gamma_{el} \approx 40$), while below ~4 K a crossover to a pronounced negative contribution takes place. The negative contribution is associated with (short-range) antiferromagnetic order ($\Gamma_{af} = -32$). The temperature variation of α of $CeInCu_2$ bears a close resemblance to that of $CeAl_3$.

1. Introduction

The cubic Heusler alloy $CeInCu_2$ has attracted considerable attention because of its classification as a heavy-fermion compound on the verge of magnetic order. In a first series of experiments the low-temperature anomalies, observed in the magnetic, transport and thermal properties [1–4], were interpreted as arising from pronounced Kondo lattice effects with a Kondo temperature $T_K \approx 6\,K$. In particular, the broad hump in the electronic specific heat centered at 2.3 K was explained as to originate from strong antiferromagnetic intersite correlations yielding an anomalously large value of the coefficient of the linear term: $\gamma = 1.2\,J/mol\,K^2$. Subsequently, on the microscopic level, NMR measurements [5,6] evidenced an antiferromagnetic transition near $T = 2\,K$. Elastic neutron-scattering experiments [7,8] confirmed antiferromagnetic order (the magnetic structure is the so-called type 1 structure of an FCC antiferromagnet) with a Néel temperature $T_N = 2.3\,K$. The ordered moment is rather small (0.3–$0.4\mu_B$/Ce-atom), a general feature of heavy-fermion antiferromagnets. However, the interpretation of the low-temperature magnetic properties is substantially complicated, because the major part of the magnetic signal appears as diffuse magnetic scattering and only a small part appears as Bragg peaks [7,8]. The diffuse magnetic scattering can be interpreted as originating from either short-range antiferromagnetic correlations or imperfect magnetic order. Indeed, the residual resistance values of the $CeInCu_2$ samples produced so far are rather large ($\rho_0 \approx 70\,\mu\Omega\,cm$) [1–4], which suggest imperfect crystalline and magnetic order. On the other hand, by substituting Ag for Cu, the residual resistance value *decreases*, while T_N increases [1] as a result of the weakened hybridization, in accordance with Doniach's phase diagram. This suggests that $CeInCu_2$ is indeed on the verge of magnetic order.

In the course of an investigation of the volume anomalies that accompany magnetic instabilities in heavy-fermion compounds (e.g., UPt_3, $CeCu_6$ and $CeRu_2Si_2$) [9,10], we here report on an accurate dilatometry study of polycrystalline $CeInCu_2$ and $LaInCu_2$ at low temperatures.

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2. Experimental

Polycrystalline $CeInCu_2$ and $LaInCu_2$ samples were shaped by means of spark erosion into a cube (edge 5 mm), with at least two surfaces parallel to within 5 μm. The samples were mounted in a three-terminal parallel-plate capacitance cell machined of oxygen-free high-conductivity (OFHC) copper [11]. The sensitive dilatometer, with a detection limit of 0.01 Å, was attached via a heat impedance to the cold plate of a ^3He cryostat operated with an adsorption pump. The coefficient of linear thermal expansion, $\alpha = L^{-1}\,dL/dT$, was measured by stepwise increasing the cell temperature ($\Delta T \geq 20\,mK$), while recording the change in length (ΔL). The measurements were performed in the temperature interval $0.3\,K < T < 12\,K$. The data have been corrected for the so-called cell effect, i.e., the signal of the cell with a dummy OFHC copper sample.

3. Results

The experimental results are shown in fig. 1. The temperature variation of α of $CeInCu_2$ is highly anomalous. It can be attributed almost completely to the anomalous behaviour of the f-electrons, since the α of the non-f-electron analog $LaInCu_2$ is at least one order of magnitude smaller in the whole temperature range. At high temperatures, α is large and positive with a maximum centered at $T_{max} = 4.5$ K. For $T < T_{max}$ a broad crossover to a pronounced negative contribution with a minimum centered at $T_{min} = 1$ K takes place. For comparison we have plotted in fig. 2 the specific heat data, $c(T)$ [1].

Recently, two other studies of $\alpha(T)$ of $CeInCu_2$ have been reported. Oomi et al. [12] employed a strain-gauge technique, focusing on a different temperature interval ($T > 4.2$ K) and, consequently, their data have little overlap with ours. A maximum in the magnetic contribution to α, observed by Oomi et al. [12] around 25 K, was interpreted as arising from a Schottky anomaly caused by the population of the first excited doublet crystal field state at ~60 K. However, the data in fig. 1 suggest that the actual analysis is more complicated because of the broad large magnetic contribution at low temperature. Matsui et al. [13] have measured $\alpha(T)$ in the temperature interval $2\,K < T < 12$ K with a capacitance technique. Their results do agree qualitatively with our data in fig. 1, but show a large scatter.

4. Discussion

First it is of interest to compare the thermal expansion and specific heat data (figs. 1 and 2). The increase

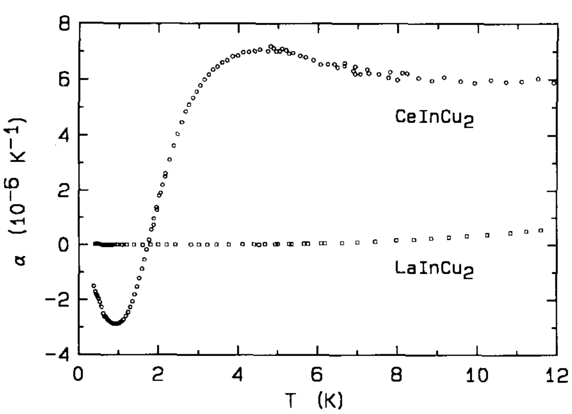

Fig. 1. Coefficients of linear thermal expansion of polycrystalline $CeInCu_2$ (○) and $LaInCu_2$ (□).

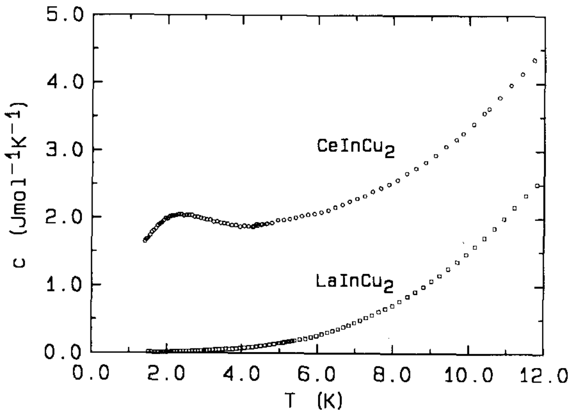

Fig. 2. Specific heat of polycrystalline $CeInCu_2$ (○) and $LaInCu_2$ (□) after ref. [1].

in $c(T)$ below $T = 4.2$ K leading to the broad hump centered at $T = 2.3$ K, concurs with a decrease in $\alpha(T)$ below $T = 4.5$ K and a maximum in $d\alpha/dT$ close to 2 K. We attribute the broad low-temperature anomaly to the above-mentioned (short-range) antiferromagnetic order [5–8].

Next we investigate the (temperature-dependent) effective Grüneisen parameter, $\Gamma_{eff}(T) = \alpha_v(T)V_m/\kappa c(T)$, where the coefficient of volume expansion is given by $\alpha_v = 3\alpha$ (cubic symmetry), $V_m = 4.728 \times 10^{-5}\ m^3/mol$ is the molar volume and $\kappa = V^{-1}\, dV/dP$ is the isothermal compressibility. Several values for κ can be found in the literature: 1.35 $Mbar^{-1}$ (from the initial pressure dependence of the lattice parameter at room temperature as determined by strain gauges up to 15 kbar [3]), 1.8 $Mbar^{-1}$ (from low-temperature elastic constants [13]) and 1.0 $Mbar^{-1}$ (from the overall pressure dependence of the lattice parameter at room temperature as determined by X-ray measurements up to 140 kbar [14]). In fig. 3 we show the electronic Grüneisen parameter $\Gamma_{el}(T) = \alpha_{v-el}(T)V_m/\kappa c_{el}(T)$ (where we used a value for κ of 1.35 $Mbar^{-1}$ [3]). The electronic contributions to $\alpha_v(T)$ and $c(T)$ have been deduced by subtracting the phonon parts, as determined by measurements on $LaInCu_2$, from the total signal. Note that in the temperature interval $0.3\,K < T < 1.3$ K, $c_{el}(T)$ has been obtained by an extrapolation conform the data in ref. [4]. Above 4 K, Γ_{el} is only weakly temperature dependent and a large value, typical of heavy-fermion compounds [9], is observed. At $T = 4$ K, $\Gamma_{el} = 42$, in good agreement with previous estimates [3,13]. The electronic Grüneisen parameter for the low-temperature contribution equals −32. Associating $\Gamma_{el} = -32$ ($T \to 0$) with the (short-range) antiferromagnetic order we deduce a suppression of T_N at

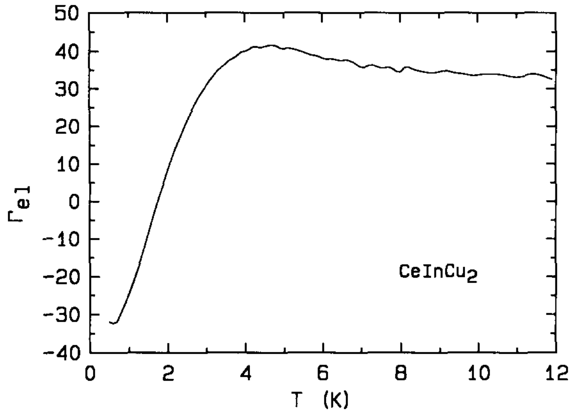

Fig. 3. The electronic Grüneisen parameter of $CeInCu_2$ (deduced with $\kappa = 1.35\,\mathrm{Mbar}^{-1}$).

a rate of $dT_N/dp = -0.1$ K/kbar. This is in good agreement with the value for $\Gamma_{af} = -35 \pm 5$ evaluated from the variation of T_N at small negative chemical pressures in the series $CeInAg_{2-x}Cu_x$ for $1.7 < x < 1.9$ [1].

The thermal expansion of $CeInCu_2$ bears a close resemblance to $\alpha(T)$ of heavy-fermion $CeAl_3$, although in the latter compound the energy scale is reduced and $\Gamma_{el}(T \rightarrow 0) = -200$ [15]. Also in $CeAl_3$ antiferromagnetic order with reduced moments develops below $T_N = 0.7$ K [16]. Furthermore, negative thermal expansion contributions below T_N have been observed in the heavy-fermion antiferromagnets $CeRu_2Si_2$ doped with La and UPt_3 doped with Pd [10]. The present data on $CeInCu_2$ confirm that the development of antiferromagnetic order in a strongly correlated electron system is accompanied by a strong reduction of the coefficient of volume expansion, in accordance with decreasing hybridization.

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