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Thermal expansion of $CeCu_{6-x}Au_x$

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Abstract

$CeCu_{6-x}Au_x$ orders antiferromagnetically for $x > 0.1$. The ordering wave vector changes drastically between $x = 0.4$ and 0.5, while the Néel temperature $T_N(x)$ varies linearly between $x = 0.1$ and 1.0. The linear thermal expansion coefficient α shows for $x = 0.3$ a positive jump $\Delta\alpha_a$ while $\Delta\alpha_b$ and $\Delta\alpha_c < 0$. When increasing x to $x = 0.5$, $\Delta\alpha_a$ and $\Delta\alpha_b$ change sign, apparently reflecting the underlying change of the magnetic ordering wave vector.

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While $CeCu_6$ is a heavy-fermion system without magnetic order, except perhaps at ultra-low temperatures ($T \sim 3$ mK), $CeCu_{6-x}Au_x$ is an incommensurate antiferromagnet for $x > x_c \approx 0.1$, with the Néel temperature T_N varying linearly in the range $0.1 < x \leqslant 1$ [1]. The latter sample, i.e. $CeCu_5Au$ with $T_N = 2.2$ K, is in fact a stoichiometric compound with the Au atoms sitting exclusively at the Cu(2) site of the $CeCu_6$ structure [2]. For $x > 1$ T_N decreases again. The ordering wave vectors determined from elastic neutron scattering reside within the reciprocal a^*c^* plane, with however a drastic change between $x = 0.4$ and 0.5 [3]. (Here we use the orthorhombic notation to denote the lattice vectors, neglecting the small monoclinic distortion by $\sim 1.5°$ for $CeCu_6$ which is rapidly suppressed with increasing x and vanishes at $x = 0.14$ [4].) For $x = 0.5$ and 1 the ordering wave vector $\mathbf{Q}$ lies on the $\mathbf{a}^*$-axis, i.e. $\mathbf{Q} = (0.59\,0\,0)$ for $x = 0.5$ and $\mathbf{Q} = (0.56\,0\,0)$ for $x = 1.0$. However, for $x < 0.5$, $\mathbf{Q}$ is distinctly different: $\mathbf{Q} = (0.625\,0\,0.275)$ for $x = 0.2$ and $\mathbf{Q} = (0.62\,0\,0.253)$ for $x = 0.3$. A similar $\mathbf{Q}$ vector was found for $x = 0.4$ [5]. We limit the discussion to long-range order with resolution-limited Bragg peaks and do not consider short-range order effects [3]. The drastic change of $\mathbf{Q}$ between $x = 0.4$ and 0.5 (see inset of Fig. 1) contrasts with the smooth linear evolution of $T_N(x)$ and presents a major puzzle. Furthermore, while hydrostatic pressure leads to a decrease of T_N for all x [1,6], uniaxial pressure measurements performed on $x = 0.2$ single crystals indicate a decrease of T_N for uniaxial pressure parallel to the **b** and **c** axes but, surprisingly, an increase for uniaxial pressure parallel to the **a**-axis [7,8]. Here we report thermal expansion measurements to shed more light on these issues.

The thermal expansion coefficient α_i ($i = \mathbf{a}, \mathbf{b}, \mathbf{c}$) was measured between 0.35 and 10 K with a parallel-plate capacitance dilatometer in a ^{3}He cryostat. The absolute error in α_i is about $3 \times 10^{-7}\,\mathrm{K}^{-1}$, which is mainly attributed to small differences in the effective area of the capacitor plates between different runs. The error in the volume expansion $\alpha_V = \alpha_a + \alpha_b + \alpha_c$ amounts to $5 \times 10^{-7}\,\mathrm{K}^{-1}$.

Figs. 1 and 2 show the thermal expansion coefficients α_i versus T for $x = 0.3$ and $x = 0.5$, respectively. The discontinuities $\Delta\alpha_i$ at $T_N = 0.5$ and 1 K, respectively, are clearly visible, although the lower measuring temperature limit of 0.35 K makes the determination of the discontinuity for $x = 0.3$ somewhat uncertain. The main

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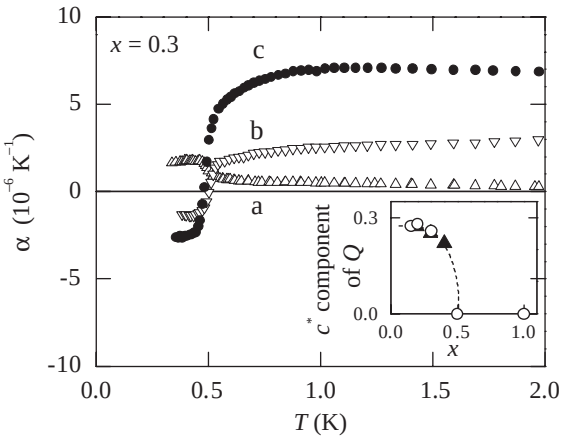

Fig. 1. Temperature dependence of the linear thermal expansion coefficients α_i (with $i = \mathbf{a}, \mathbf{b}, \mathbf{c}$) of $CeCu_{5.7}Au_{0.3}$. The inset shows the reciprocal $\mathbf{c}^*$-axis component of the magnetic ordering wave vector $\mathbf{Q}$.

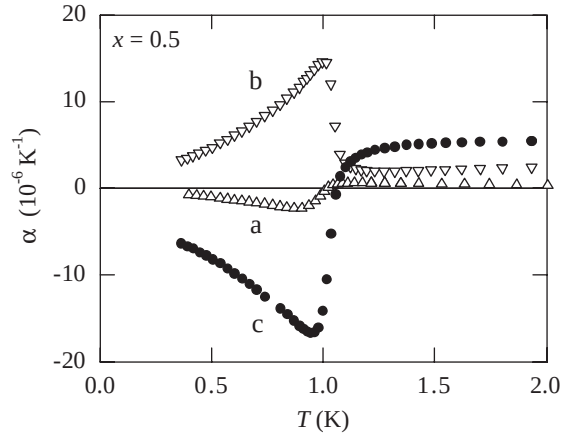

Fig. 2. Temperature dependence of the linear thermal expansion coefficients of $CeCu_{5.5}Au_{0.5}$.

differences between $x = 0.3$ and 0.5 are (i) the magnitude of $\Delta\alpha_i$ and (ii) the sign of $\Delta\alpha_i$. Concerning (i), $\Delta\alpha_V = \Delta\alpha_s + \Delta\alpha_b + \Delta\alpha_c$ increases roughly by a factor 3 between $x = 0.3$ and 0.5. This increase matches nicely the increase of the specific-heat discontinuity ΔC_p [3]. Since the ratio $\Delta\alpha_V/\Delta C_p$ can be related to the hydrostatic pressure dependence of T_N, dT_N/dp, by the Ehrenfest relation

$$dT_N/dp = T_N V_{mol} \Delta\alpha_V/\Delta C_p \quad (1)$$

the approximately concordant behavior of both discontinuities indicates a slowly varying dT_N/dp. In fact, the obtained values of about -0.7 K/GPa for $x = 0.3$ and -0.4 K/GPa for $x = 0.5$ are in reasonable agreement with the directly measured dT_N/dp values [6].

Concerning (ii), we note that while $\Delta\alpha_c$ is negative for both $x = 0.3$ and $x = 0.5$, $\Delta\alpha_a > 0$ and $\Delta\alpha_b < 0$ for $x = 0.3$, but $\Delta\alpha_a < 0$ and $\Delta\alpha_b > 0$ for $x = 0.5$. The data for $x = 0.3$ are thermodynamically consistent with previously unexpected $dT_N/d\sigma_a > 0$, while $dT_N/d\sigma_b$ and $dT_N/d\sigma_c < 0$ as determined from measurements of the shift of the specific-heat maximum at T_N under uniaxial pressure σ_i for a $x = 0.2$ crystal [7]. By applying the Ehrenfest relation (1) to each individual $\Delta\alpha_i$, we get for $x = 0.3$, $dT_N/d\sigma_i = 0.07$, -0.21, and -0.56 K/GPa, while for $x = 0.5$, $dT_N/d\sigma_i = -0.1$, 0.39, and -0.67 K/GPa along the **a**-, **b**-, and **c**-axis, respectively. These uniaxial stress dependences for $x = 0.3$ deviate from the specific-heat derived values for the $x = 0.2$ compound, probably because of friction losses in the particular pressure cell used for the specific-heat measurements [7]. A comparison with a careful resistivity measurement under uniaxial stress along the **a**-axis of a $x = 0.2$ crystal [8] does reveal with -0.065 K/GPa the same value of $dT_N/d\sigma_a$.

The change of the uniaxial pressure dependence between $x = 0.3$ and 0.5 is quite likely linked with the change of the magnetic ordering wave vector, although a direct explanation is not at hand. The change is clearly not related to a simple volume effect, since the variation of lattice parameters with x is linear up to $x = 1$ [6]. It is remarkable, however, that while the overall volume increases (of course also linearly) with x, it is only the **a** and **c** axes that expand, while the **b**-axis shrinks a little. We are left with the ad-hoc assumption that due to the delicate balance of RKKY interaction and Kondo screening, which is strongly dependent on interatomic distances, the RKKY interaction is favored by expanding the lattice along **b** and **c** for $x = 0.3$, corresponding to an increase of T_N with interatomic distance, and along **a** and **c** for $x = 0.5$. It is interesting to note that above T_N, the thermal expansion for both alloys is almost identical, suggesting that the Kondo effect does not vary too strongly between $x = 0.3$ and 0.5. Elastic neutron scattering experiments under pressure are underway to elucidate the magnetic structure and to check whether a reorientation of $\mathbf{Q}$ occurs when a $x = 0.5$ sample is compressed to a volume corresponding to $x = 0.3$.

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