Thermal expansion of monocrystalline heavy-fermion $CeCu_6$

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(Received 27 December 1988)

Thermal-expansion measurements on monocrystalline $CeCu_6$ in the temperature range $1.5 < T < 100$ K are presented. The coefficient of linear thermal expansion is strongly anisotropic. The volume expansion exhibits a maximum at 2.5 K that is attributed to the Kondo-lattice effect. The data are compared with specific-heat data ($T < 20$ K) on the same sample. An analysis in terms of Grüneisen parameters yields the large value $\Gamma_{el} = 57$ for the heavy-fermion contribution.

I. INTRODUCTION

The intermetallic compound $CeCu_6$ has been widely studied[1] due to its unequaled heavy-fermion properties. The large coefficient (γ) of the linear electronic specific heat[2–4] amounts to ~ 1.6 J/mol K^2, i.e., 200 times larger than that of its nonheavy counterpart $LaCu_6$ ($\gamma = 8$ mJ/mol K^2, Ref. 3). The enhancement of the γ value arises from the presence of the $4f$ (Ce) electrons that hybridize with the Cu ligand $3d$ or $3p$ orbitals, forming a narrow quasiparticle band at the Fermi surface. Within the Fermi-liquid description the effective mass of the quasiparticles amounts to $m^* \simeq 480 m_e$ and, accordingly, the Fermi temperature (T_F) equals ~ 90 K (here we ignore anisotropy effects and use a Fermi radius[5] $k_F = 10^{10}$ m^{-1}). No evidence for superconductivity or long-range magnetic order has been found[6] down to temperatures as low as ~ 18 mK. The apparent lack of an ordered ground state renders $CeCu_6$ comparable to the archetype heavy-fermion compound $CeAl_3$ ($\gamma \simeq 1.6$ J/mol K^2, Ref. 7), which is furthermore supported by similarities[8] in the low-temperature thermal and transport properties. However, the compound $CeCu_6$ offers the advantage that fairly large single-crystalline samples can be grown.[9] This makes a study of its strongly anisotropic properties (orthorhombic structure) feasible.

The specific heat of $CeCu_6$ has first been measured by Stewart *et al.*[2] ($1.8 < T < 38$ K) and Fujita *et al.*[3] (150 mK $< T <$ 70 K). The latter authors also separated the electronic contribution by subtracting the lattice contribution obtained on $LaCu_6$. The resulting specific heat was found to exhibit two contributions centered at 2.6 and 27 K. Under the orthorhombic symmetry the crystal field splits the $J = \frac{5}{2}$ level into three doublets. The low-temperature anomaly has been attributed[3] to the Kondo effect on the doublet ground state, with $T_K = 3.9$ K. The second anomaly has been attributed[3] to a Schottky anomaly caused by the population of the first excited doublet at $\Delta_1/k_B = 65$ K. The entropy near 70 K amounts to $\sim R \ln 4$. Careful specific-heat measurements ($20 < T < 220$ K), performed by Felten,[10] indicate that the magnetic entropy approaches $R \ln 6$ at 220 K. This suggests that the energy separation between the excited doublets is rather small, e.g., 40 K, leading to a level scheme 0–60–100 K. Similar crystal-field splittings have been inferred from the temperature dependence of the elastic constants[11] (0–71–117 K) and from inelastic neutron-scattering experiments[12] (0–64–128 K). However, since the excitation energies for the crystal field are rather small, one cannot neglect the Kondo effect on the excited doublet states, that in general will cause a considerable broadening of the crystal-field levels. The admixture of the Kondo and crystal-field effect leads in the model of Hanzawa *et al.*[13] to an effective Kondo temperature for the full multiplet of $T_K^h = (T_K \Delta_1 \Delta_2)^{1/3} \cong 30$ K.

Magnetic susceptibility measurements[14,15] on single-crystalline samples reveal a Curie-Weiss behavior above ~ 100 K with an effective moment close to the free-ion value of Ce^{3+} ($2.54 \mu_B$) for all three crystallographic directions. The paramagnetic Curie temperature amounts to -50 K for the a and b axis and to ~ -10 K for the c axis, indicating the presence of antiferromagnetic interactions. Anomalies in the susceptibility below 100 K have been attributed to crystal-field effects. Calculations[15] result in a level scheme 0–107–241 K, somewhat at variance with the energy splittings quoted above. Inelastic neutron-scattering experiments[16,17] have confirmed the presence of antiferromagnetic excitations in the low-temperature Fermi-liquid regime. This suggests that $CeCu_6$ is close to an antiferromagnetic instability, a conclusion also drawn for several other heavy-fermion compounds, e.g., $CeAl_3$ (Ref. 18), $CeRu_2Si_2$ (Ref. 19), and UPt_3 (Ref. 20). Indeed, by exerting negative chemical pressure $CeCu_6$ exhibits long-range antiferromagnetic order, as was recently shown by alloying experiments on $CeCu_{6-x}Au_x$ (Ref. 21) and $CeCu_{6-y}Ag_y$ (Refs. 21 and 22) ($T_N = 0.95$ K for $x = 0.5$, $T_N = 0.58$ K for $y = 0.6$).

Thermal expansion and specific heat are related to each other by means of Grüneisen parameters. Recently, it

has been shown that the electronic Grüneisen parameter for the heavy-fermion liquid, $\Gamma_{el} = -\partial \ln T_F / \partial \ln V$, exceeds the free-electron value by two orders of magnitude, e.g., Γ_{el} amounts to -200, 70, and 150 for $CeAl_3$ (Ref. 23), UPt_3 (Ref. 24), and $CeRu_2Si_2$ (Ref. 25), respectively. This implies that the thermal expansion is more sensitive to the heavy-fermion behavior than the specific heat itself. Besides, a combination of thermal-expansion and specific-heat data allows for the determination of the volume dependence of the characteristic temperatures and may serve as a check of their direct measurements as function of hydrostatic pressure. Furthermore, thermal expansion may be used to study anisotropy effects, in contrast to specific heat which is a bulk property.

In this contribution we report on thermal-expansion measurements on a single-crystalline sample in the temperature range $1.5 < T < 100$ K. The data will be compared with specific-heat measurements[26,27] on the same sample ($T < 20$ K). Earlier thermal-expansion measurements were performed by Oomi *et al.*[28] ($6 < T < 200$ K), employing a strain-gauge technique. After correcting for the lattice expansion, Oomi *et al.*[28] analyzed the resulting contribution in a crystal-field model with level scheme 0–35–104 K. When analyzing our data, we shall focus on the low-temperature heavy-fermion contribution. In addition we studied two polycrystalline samples: a pure $CeCu_6$ sample and a sample in which part of the Cu has been substituted by Al, with composition $CeCu_{5.75}Al_{0.25}$.

II. EXPERIMENT

$CeCu_6$ crystallizes in the orthorhombic structure (space group D_{2h}^{16}) but undergoes a phase transition to a monoclinic structure[29,30] (space group C_{2h}^5) at about 220 K. Since the monoclinic distortion is rather small ($\beta = 91.5°$ at 4.2 K, Ref. 30), we retain the orthorhombic notation for the crystal axes. The single-crystalline sample has been pulled out of a tungsten crucible, employing the Czochralski technique.[9,31] Starting materials were 99.99% pure Ce (Rare Earth Products Ltd.) and 99.999% pure Cu (Koch Light Ltd.) No additional annealing has been performed. The lattice parameters[8,31] amount to $a = 8.104$, $b = 5.106$, and $c = 10.175$ Å. The residual resistance for this crystal[8,31] equals 15.7, 8.3, and 9.2 $\mu\Omega$ cm for the a, b, and c axis, respectively.

The coefficient of linear thermal expansion $\alpha = (1/L)(dL/dT)$, has been measured using a sensitive three-terminal capacitance method, with a detection limit for length changes of 0.1 Å. Hereto the sample was shaped into a parallelepiped by means of spark erosion and placed in a capacitance cell machined of oxygen free high-conductivity copper.[32] The sample edges were taken (within $\sim 1°$) along the main crystallographic directions. The dimension of the sample equals $3.8 \times 5.0 \times 3.8$ mm^3. Data were gathered stepwise, with temperature steps varying between 0.2 K in the low-temperature range and ~ 3 K near 100 K. The accuracy in α is limited to $\sim 3 \times 10^{-7}$ K^{-1} at 100 K due to small temperature gradients over the cell. Below 50 K temperatures were read with a carbon-glass thermometer and there above with a platinum resistor.

Polycrystalline $CeCu_6$ and $CeCu_{5.75}Al_{0.25}$ were prepared by arc melting. The samples were shaped by means of spark erosion into a cube with an edge of 5 mm.

III. RESULTS

The coefficients of linear thermal expansion α_a, α_b, and α_c for single-crystalline $CeCu_6$ are shown in Fig. 1(a) for temperatures below 100 K. The low-temperature part is shown in detail in Fig. 1(b). Below 15 K, the data consist of several runs that have been taken with increasing temperature. Above 15 K one single run was made. A measurement of the a axis in the low-temperature region with decreasing temperature revealed some hysteresis effects (in the order of $\sim 5 \times 10^{-7}$ K^{-1}). These are possibly caused by the friction between the surface of the sample that contains the bc plane and the copper cell (due to the large mismatch of expansion coefficients). For the other crystallographic directions hysteresis effects have not been observed. Relative length changes $\Delta L/L$ with respect to 1.5 K have been obtained by integrating the α versus T curves. The results are displayed in Fig. 2. The present data for the b and c axis are in good agreement with earlier data ($T > 6$ K) of Oomi *et al.*[28] obtained with

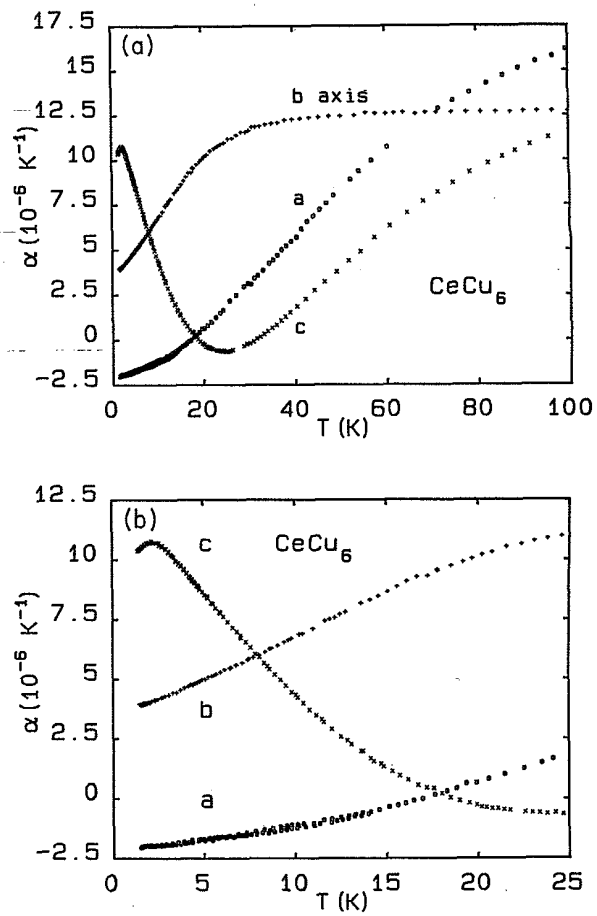

FIG. 1. Coefficient of linear thermal expansion for single-crystalline $CeCu_6$ (a) below 100 K and (b) below 25 K.

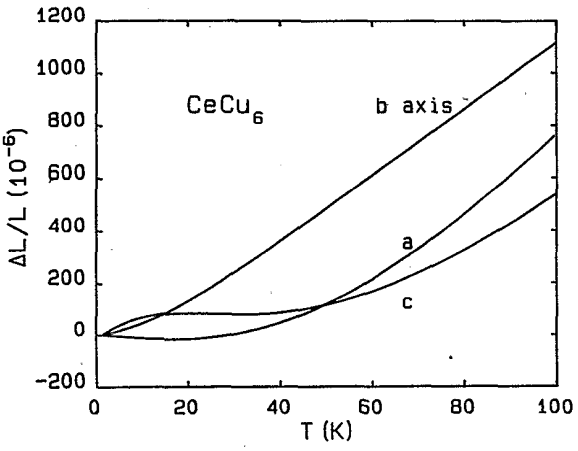

FIG. 2. Relative length change with respect to 1.5 K for single-crystalline $CeCu_6$. Crystallographic directions as indicated.

the strain-gauge technique. Data for the a axis have not been published so far.

The volume effect has been calculated from $\alpha_v = \alpha_a + \alpha_b + \alpha_c$, after smoothing the data for α_a, α_b, and α_c in selected temperature intervals by polynomial fits. The resulting values for $\alpha_v/3$ are shown in Fig. 3, together with the data for polycrystalline $CeCu_6$ and $CeCu_{5.75}Al_{0.25}$.

IV. ANALYSIS AND DISCUSSION

As follows from Figs. 1 and 2 the thermal expansion of $CeCu_6$ is strongly anisotropic. With raising temperature the unit cell expands along the b and c axes, while it shrinks along the a axis up to 17 K. However, above 19 K the c axis begins to shrink, till it expands again above 31 K. This complex behavior is reflected in the various extrema observed in the volume expansion (Fig. 3).

When comparing the α_v curves for the single and polycrystalline $CeCu_6$ samples we note the somewhat weaker temperature dependence and the absence of the low-temperature maximum for the latter sample. One has to realize, however, that the samples are of different sources, and that preferred alignment of the crystallites might cause a slightly anisotropic dilatation of the polycrystalline cube that has been measured along one edge only. In the following analysis we will concentrate on the results for the single-crystalline sample.

In order to derive the electronic contribution (α_{el}) to the thermal expansion of $CeCu_6$, the phonon part (α_{ph}), obtained from, for example, measurements on isostructural $LaCu_6$, need to be subtracted (the conduction-electron contribution to the expansion coefficient of $LaCu_6$ can readily be neglected). Measurements of single-crystalline $LaCu_6$ along the three crystallographic directions have been performed by Oomi *et al.*,[28] revealing a substantial anisotropy. From their data we have calculated the coefficient of volume expansion α_v which is shown as a dashed line in Fig. 3.

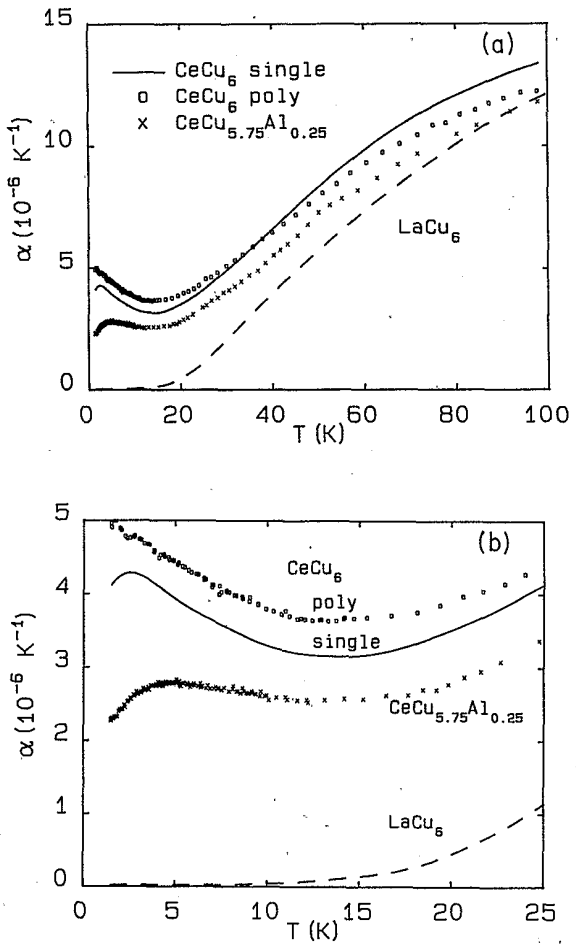

FIG. 3. One-third of the volume expansion ($\alpha_v/3$) for single-crystalline (solid line) and polycrystalline (○) $CeCu_6$, polycrystalline $CeCu_{5.75}Al_{0.25}$ (×) (a) below 100 K and (b) below 25 K. The dashed line indicates the phonon contribution represented by $LaCu_6$ (Ref. 28).

The phonon-corrected thermal expansion of $CeCu_6$ is shown in Fig. 4. Two contributions can be discerned: (1) a pronounced low-temperature anomaly centered at 2.5 K that we attribute to the heavy-fermion behavior, and (2) a broad high-temperature contribution with a maximum (that appears after separating out the heavy-fermion contribution) at ~60 K, associated with the crystal-field effects (α_{cf}). In Fig. 5 we show the corresponding electronic specific heat. Below 30 K this curve has been constructed by correcting the specific heat of $CeCu_6$, measured by Fisher *et al.*[26] on the same sample as used for the present experiments, with the $LaCu_6$ data of Fujita *et al.*[3] Above 20 K we reproduced the data of Felten.[10] Note that the error bar in the high-temperature region is large (1.5 J/mol K), due to the relatively small difference of the specific heats of $CeCu_6$ and $LaCu_6$ (see also Ref. 33). Here the low-temperature contribution is centered[3,33] at ~2.5 K, while the crystal-field contribution (c_{cf}) leads to a maximum at 27 K.

Comparing Figs. 4 and 5, it is remarkable that α_{cf} is centered at a considerably higher temperature than c_{cf}.

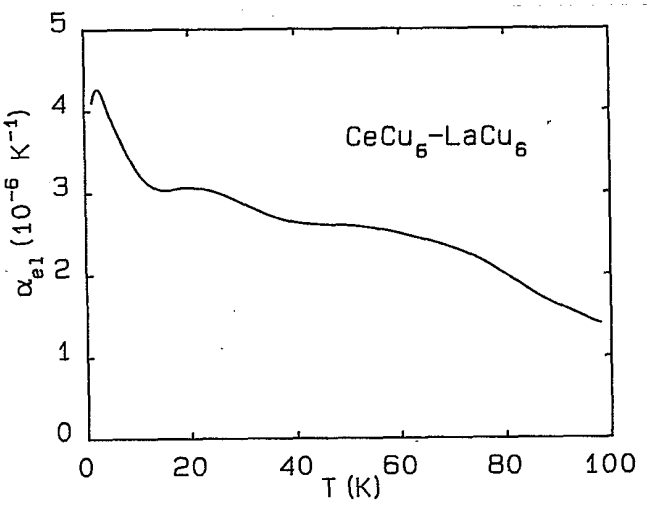

FIG. 4. Electronic contribution α_{el} to the thermal expansion of $CeCu_6$.

A reason for this might be the nearness of the excited crystal-field levels as follows from the level scheme[12] 0–64–128 K. The presence of two different Grüneisen parameters $\Gamma_{cf}^{1,2}=-(\partial \ln\Delta_{1,2}/\partial \ln V)$ for the two crystal-field splittings (Δ_1 and Δ_2) might furthermore lead to different magnitudes of the crystal-field contributions in the specific heat and thermal expansion and hence to different temperatures at which the extrema in α and c occur. Oomi *et al.*[28] concluded that the crystal field mainly contributes to the expansion along the b axis. From a crystal-field fit to $\alpha_b(CeCu_6)$-$\alpha_b(LaCu_6)$ these authors derived the level scheme 0–35–104 K. In particular the lowest splitting is too small when compared with the analysis of the specific heat. We shall comment on this point later.

In order to separate the various contributions to the thermal properties, an analysis in terms of Grüneisen parameters can be performed. One has to bear in mind, however, that such an analysis may only serve as a first approximation due to the forementioned admixture of the crystal-field and Kondo effect. We define an effective temperature-dependent Grüneisen parameter $\Gamma_{eff}=(\alpha_v V_m/\kappa c)$, where α_v and c are the total volume thermal expansion and specific heat, respectively, V_m is the molar volume, and κ is the isothermal compressibility. In general, if the effective Grüneisen parameter is constant in a certain temperature interval, one can identify Γ_{eff} with the Grüneisen parameter for the particular mechanism that governs the thermal properties in that temperature range. Such an analysis has been applied successfully to the heavy-fermion compounds UPt_3 (Ref. 34) and $CeRu_2Si_2$ (Ref. 25). In the latter compound, for example, the spin-fluctuation contribution (centered at 8 K) and the crystal-field contribution (centered at 90 K) could be separated employing the Grüneisen parameters $\Gamma_{sf}=150$ and $\Gamma_{cf}=-4$.

The effective Grüneisen parameter for $CeCu_6$ is shown in Fig. 6, up to 100 K. Here we used $V_m=6.33\times10^{-5}$ m^3/mol and $\kappa=1.21$ $Mbar^{-1}$. The value for κ has been derived from the pressure dependence of the (020), (104), and (311) lattice spacings, measured by Shibata *et al.*[35] The compressibility is strongly anisotropic. The linear compressibilities amount to $\kappa_a=-1/a(da/dp)=0.14$ $Mbar^{-1}$, $\kappa_b=0.66$ $Mbar^{-1}$, and $\kappa_c=0.41$ $Mbar^{-1}$, from which it follows $\kappa=\kappa_a+\kappa_b+\kappa_c=1.21$ $Mbar^{-1}$. In the low-temperature range Γ_{eff} is strongly temperature dependent, extrapolating to the large value of 57 at 0 K. In the high-temperature part, where α_v and c are dominated by phonons, $\Gamma_{eff}=1.7$, close to the value $\Gamma_{ph}=2$ observed for normal metals. The insert in Fig. 6 shows the electronic Grüneisen parameter, $\Gamma_{el}=(3\alpha_{el}V_m/\kappa c_{el})$, up to 25 K. After a sharp drop at low temperature, Γ_{el} levels off near 15 K and is more or less temperature independent up to 100 K, with a mean value of 14. The large overlap of the heavy-fermion and crystal-field con-

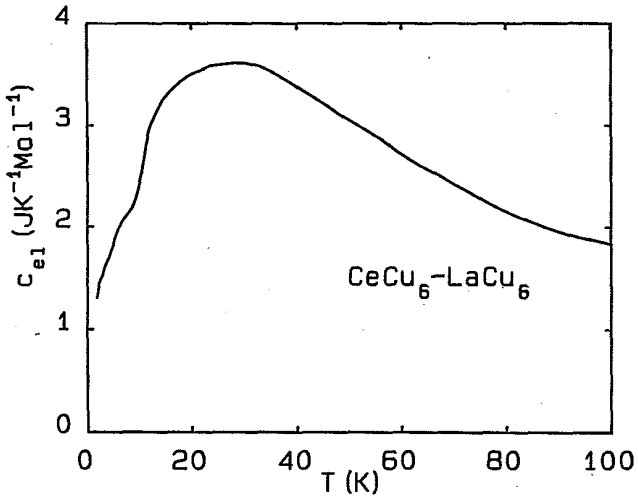

FIG. 5. Electronic contribution c_{el} to the specific heat of $CeCu_6$ (Refs. 3, 10, and 26).

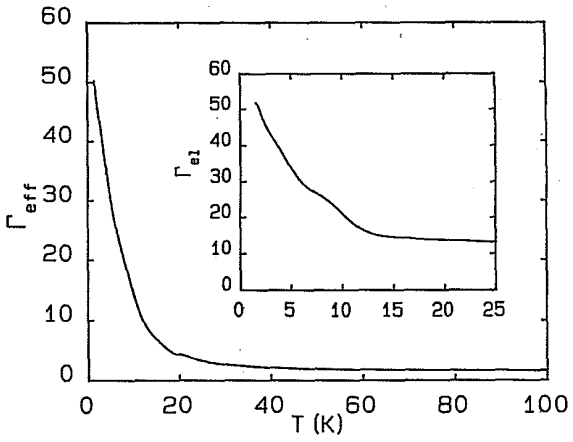

FIG. 6. Temperature dependence of the effective Grüneisen parameter for $CeCu_6$. The inset shows the electronic Grüneisen parameter up to 25 K.

tributions and, not in the least, the large error bars at high-temperature for α_{el} and c_{el}, make it not possible to determine the crystal-field Grüneisen parameters unambiguously from the Γ_{el} versus T curve. However, the Γ_{cf}'s are undoubtedly positive. A crude estimate for Γ^1_{cf} for the lowest crystal-field splitting amounts to $\Gamma^1_{cf}=13$. This implies that Δ_1/k_B increases with pressure at a rate of ~ 1 K/kbar. Unfortunately, information on the pressure dependence of Δ_1 (or Δ_2) from $c(p)$, $\chi(p)$, magnetostriction, or other experiments is not available yet.

Next we focus on the low-temperature anomaly. For heavy-fermion compounds one usually approximates the low-temperature specific-heat and expansion coefficient by $c=\gamma T$ and $\alpha_v=3aT$, respectively. As follows from Fig. 3(b), the linear term is not attained yet. At 1.5 K, $\alpha_v/3T$ amounts to 2.75×10^{-6} K^{-2}, but extrapolates to $a\simeq 6\times10^{-6}$ K^{-2} at 0 K. Similarly, c/T amounts to 0.853 J/mol K^2 at 1.5 K, but reaches a value[26] of $\gamma=1.67$ J/K^2 mol at 0 K. The resulting low-temperature electronic Grüneisen parameter, $\Gamma_{el}=(3aV_m/\kappa\gamma)$, equals 57 (see also Fig. 6). Knowing Γ_{el}, one can calculate the pressure dependence of γ,

$$\frac{d\gamma}{dp}=-\kappa\Gamma_{el}\gamma=-114\ (\text{mJ/mol K}^2)/\text{kbar}\ ,$$

a value in good agreement with the one calculated directly from the $c(p)$ experiments,[26] -145 (mJ/mol K^2)/kbar. In the Kondo model Γ_{el} can be used to determine the pressure dependence of T_K, since $\gamma=0.68R/T_K$ (Ref. 36), where $T_K=T_{max}/0.66$ (T_{max} is the temperature at which the maximum in the specific heat occurs) and R is the gas constant and thus

$$\Gamma_{el}=-\frac{\partial \ln T_K}{\partial \ln V}\ .$$

With $T_K=3.9$ K, we derive

$$\frac{dT_K}{dp}=\kappa\Gamma_{el}T_K=0.27\ \text{K/kbar}\ .$$

Transport measurements yield a similar value for Γ_{el}: From the initial ($p<5$ kbar) pressure dependence of the maximum in the resistivity,[35,37] $(dT_{max})/dp=1$ K/kbar, it follows, under the assumption $T_{max}(\cong 15\ \text{K})\propto T_K$, that $-(\partial \ln T_K/\partial \ln V)=56$.

An alloying $CeCu_6$ with Al the low-temperature anomaly broadens and shifts towards higher temperatures (Fig. 3). When replacing 4.17% of the Cu by Al the temperature at which the maximum in α occurs equals 4.7 K versus 2.6 K for pure $CeCu_6$. In the Kondo model we subsequently derive $T_K=7.1$ K and $\gamma=0.8$ J/mol K^2. Extrapolating the α versus T curve to 0 K, we derive $a\simeq 3.2\times10^{-6}$ K^{-2} and, combining specific heat and thermal expansion, $\Gamma_e=64$. This indicates that Γ_{el} does not change much at low-Al contents. The increase of T_K can be easily understood as caused by the (positive) chemical pressure induced by alloying. Also for $CeRu_2Si_2$, $\Gamma_{el}\simeq 150$ was found to be volume independent, for relatively small volume changes ($\Delta V/V\sim 0.01$), as followed from alloying studies with La and Y.[38] Substitution of La for Ce[39] and Au or Ag for Cu[21,22] in $CeCu_6$ results in a negative chemical pressure. In the pseudobinary compounds $Ce_{1-x}La_xCu_6$ the unit cell expands (mainly along the c axis) with increasing La contents. From the lattice parameters tabulated in Ref. 39 we calculate $\Delta V/V\sim 0.035$ for $x=0.5$. Assuming Γ_{el} volume independent, we deduce a depression of T_K of $\Delta T_K/T_K=-0.20$ (for $x=0.5$). From resistivity measurements along the b axis Onuki *et al.*[33] claim, however, that T_K is nearly independent of the Ce concentration, which puts some question marks as to the model in which these authors determined T_K. In the case of $CeCu_{6-x}Ag_x$ a volume expansion $\Delta V/V\sim 0.005$ for $x=0.6$ has been reported.[40] Again, assuming $\Gamma_{el}\simeq 60$, one calculates $\Delta T_K/T_K=-0.30$. The measured γ value (for $x=0.6$) amounts to 2.8 J/mol K^2 implying $\Delta T_K/T_K=-0.46$, a 50% larger value. However, recently it has been reported[22] that $CeCu_{5.4}Ag_{0.6}$ orders magnetically at 0.58 K, which makes the determination of T_K from the available low-temperature specific-heat data not unambiguous.

In the last section of this paper we address the large anisotropy in the thermal expansion coefficient. Above ~ 20 K the anisotropy is a result of the anisotropic phonon contribution and the crystal-field effect. According to Oomi *et al.*[28] the crystal field contributes mainly along the b axis. However, it is likely that the minimum near 25 K along the c axis is also a result of the crystal-field splitting. Therefore, the maximum in α_{el} (at ~ 60 K) occurs at a higher temperature than in the phonon corrected α_b (Ref. 28). As a result, a crystal-field fit to the volume expansion will lead to a higher first excited level, in agreement with the specific heat, than a fit to α_b alone. The Kondo effect and the herewith related large value of γ show up mainly in the expansion along the c axis, i.e., the steep rise (with decreasing temperature) below 25 K, and the low-temperature maximum at 2.5 K. Below 1.5 K, α_a and α_b must have an extremum as well, since eventually α goes to 0 at 0 K. We believe that the large anisotropy at low temperatures arises from the anisotropic spin-fluctuation spectrum. Inelastic neutron-scattering experiments[17] have shown that the antiferromagnetic fluctuations are confined to the c axis, which is also the easy axis for magnetization.[14] The coupling between adjacent bc planes was found to be antiferromagnetic. Consequently, the coefficient of expansion is largest along the confinement direction α_c and smallest (negative) between the coupled planes (α_a). Also for the systems UPt_3 and $CeRu_2Si_2$ we notice a correlation between the direction of confinement of spin fluctuations and the anisotropy in the thermal expansion. Related heretofore one observes that the thermal expansion is largest for the easy direction of magnetization.

To summarize, we have measured the thermal expansion of single-crystalline $CeCu_6$. At 2.5 K a Kondo anomaly appears in the volume expansion. The heavy-fermion Grüneisen parameter amounts to 57, in agreement with similar values deduced from various pressure experiments. At low temperatures the linear thermal expansion reveals a large anisotropy. This can be understood from the anisotropic magnetic excitation spectrum.

ACKNOWLEDGMENTS

We thank A. Amato and E. Walker for preparing the single-crystalline sample and B. Barbara for supplying the polycrystalline samples. Furthermore, we thank G. Oomi for making unpublished data on $LaCu_6$ for us available. One of us (A.L.) gratefully acknowledges support from Conselho Nacional de Desenvolvimento Científico e technológico (CNPq) (Brazil). The research of A.d.V. was made possible by a fellowship of the Royal Netherlands Academy of Arts and Sciences.

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