Uniaxial Pressure Effects on $CeIrIn_5$ and $CeCoIn_5$ Studied by Low-Temperature Thermal Expansion

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We report low-temperature thermal expansion measurements on the tetragonal heavy-fermion superconductors CeMIn$_5$ (M = Ir, Co) in magnetic fields up to 8 T which allow for the analysis of the uniaxial pressure effects on both normal-state and superconducting properties. Our study reveals that T_c is strongly affected by at least two factors, the lattice anisotropy and the $4f$-conduction-electron hybridization strength which is most sensitive to c-axis lattice distortions. Non-Fermi-liquid behavior caused by quantum-critical fluctuations is observed for both systems, most pronounced for $CeCoIn_5$.

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Heavy-fermion (HF) superconductivity, first discovered in $CeCu_2Si_2$ [1] and subsequently observed in several U-based HF systems [2–4], continues to attract the interest of many researchers due to nonexponential temperature dependences of the specific heat and related properties, multiple superconducting (SC) phases, and spontaneous breaking of time-reversal symmetry [5]. These observations indicate an unconventional superconducting state with an order parameter possessing a symmetry lower than that of the underlying crystal lattice. Furthermore, the presence of a strong interaction between localized f states and itinerant conduction electrons allows for the possibility of a nonphonon mediated coupling as recently demonstrated for UPd_2Al_3 [6]. Arguments in favor of magnetically mediated coupling derive from the observation that in the Ce-based HF compounds $CeCu_2Ge_2$ [7], $CePd_2Si_2$ [8], $CeRh_2Si_2$ [9], and $CeIn_3$ [10] the SC phase exists at the border of antiferromagnetic (AF) order. In these compounds the AF ordering temperature is continuously reduced by the application of hydrostatic pressures. This defines an AF quantum-critical point (QCP) at a critical pressure p_c where $T_N \rightarrow 0$. At $p = p_c$, the low-T resistivity follows a $(\rho - \rho_0) \propto T^n$ dependence with $1 \leq n \leq 1.5$, indicative of a strong deviation from Landau-Fermi-liquid (LFL) behavior for which $(\rho - \rho_0) \propto T^2$. Additionally, in the region around p_c the onset of superconductivity is observed in the above mentioned compounds. The recently discovered AF HF compound $CeRhIn_5$ ($T_N = 3.8$ K) crystallizes in a tetragonal crystal structure which can be viewed as layers of $CeIn_3$ separated by sheets of $RhIn_2$. Near its QCP for pressures exceeding $p_c = 1.63$ GPa, $CeRhIn_5$ has been shown to become a HF superconductor below $T_c = 2.1$ K [11]. The fact that T_c of $CeRhIn_5$ exceeds that of $CeIn_3$ ($T_c = 0.2$ K at $p_c = 2.5$ GPa) by 1 order of magnitude has been attributed to the quasi-two-dimensional crystal structure [11] that leads to cylindrical Fermi surface sheets [12]. Furthermore, the quasilinear T dependence of the electrical resistivity above T_c is compatible with two-dimensional critical fluctuations at the AF QCP [11].

The two isostructural HF compounds $CeIrIn_5$ [13] and $CeCoIn_5$ [14] are ambient pressure superconductors at $T_c = 0.4$ K and 2.3 K, respectively, which are located slightly beyond the QCP on the paramagnetic side. This allows for a full thermodynamic analysis using both specific heat, C, and thermal expansion, α, results for the SC and normal-state taken at ambient pressure. The linear thermal expansion coefficient is defined as $\alpha = l^{-1}(\partial l/\partial T)$, where l denotes the sample length. As the α values of HF systems exceed those for ordinary metals by a factor 10^3–10^4 the low-T Grüneisen parameter, being proportional to the ratio α/C, is strongly enhanced. Combining the results of the thermal expansion measured along different crystalline directions with specific-heat data reveals important information on the anisotropy of the pressure dependences of both characteristic energy scales: the Kondo-lattice temperature T^* and the SC transition temperature T_c. Since the In atoms in CeMIn$_5$ reside on two nonequivalent positions—In(1) in the tetragonal plane and In(2) in the MIn_2 layer—a directional thermal expansion study can uncover the relative importance of the Ce-In(1) (in-plane) compared to the Ce-In(2) (out-of-plane) exchange interaction. In this Letter we report low-T thermal expansion measurements on single-crystalline $CeIrIn_5$ and $CeCoIn_5$ samples down to 50 mK and in magnetic fields up to 8 T. To determine $\alpha(T, B)$ an ultrahigh-resolution capacitive dilatometer with a maximum sensitivity corresponding to $\Delta l/l = 10^{-11}$ [15] was utilized. Single crystals of $CeIrIn_5$ and $CeCoIn_5$ were grown as described in [13]. They have a platelike shape with the long axis perpendicular to the c direction. The sample size is 1.8 mm along a and 1.6 mm along c for $CeIrIn_5$. The length of the $CeCoIn_5$ crystal is $l_a = 2.4$ mm

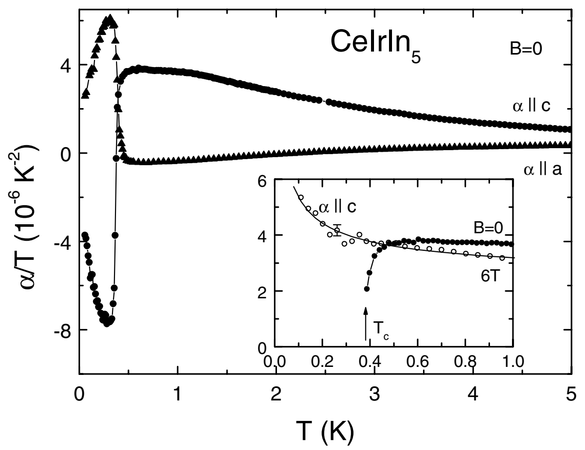

FIG. 1. Linear thermal expansion coefficients, α_a/T and α_c/T, vs T for $CeIrIn_5$. The inset shows α_c/T vs T at $B = 0$ (solid symbols) and 6 T (open symbols). The solid line describes the fit $\alpha/T = aT^{-0.5} + b$ with $a = 10^{-6}\,K^{-1.5}$ and $b = 2.19 \times 10^{-6}\,K^{-2}$ for $B = 6\,T$ below 1 K.

and $l_c = 0.5$ mm. To check for reproducibility, a second $CeCoIn_5$ single crystal has been measured along the c axis with $l = 0.75$ mm yielding identical results.

Figure 1 shows that the thermal expansion of $CeIrIn_5$ is strongly anisotropic at low temperatures [16]. In particular, the jump anomalies at $T_c = 0.38$ K, $\Delta\alpha_a$, and $\Delta\alpha_c$, have opposite signs. The values of $\Delta\alpha$ are estimated as usual by an equal-areas construction in α/T vs T plots. By using the Ehrenfest relation, $\partial T_c/\partial p_{a,c} = V_{mol} T_c \Delta\alpha_{a,c}/\Delta C$, we obtain the uniaxial pressure dependences of T_c in the zero-pressure limit. Here V_{mol} and ΔC denote the molar volume and the jump anomaly at T_c in the specific heat [13], respectively. The so-derived uniaxial pressure dependences have opposite signs (cf. Table I). The value calculated for the hydrostatic pressure dependence agrees reasonably well with that obtained by specific-heat measurements in a pressure cell [18]. Since uniaxial pressure applied along the c axis strongly reduces T_c whereas an increase of T_c is indicated for pressure applied in the basal plane, a clear relation between T_c and the lattice anisotropy can be inferred: Increasing the c/a ratio enhances T_c.

At $B = 0$, an analysis of the temperature dependence is complicated by an anomaly that occurs at around 1.2 K. This anomaly of unknown origin also appears at 1 K in an overcritical field of $B = 6\,T$ for $\alpha \parallel c$. However, in this field a pronounced increase of α/T along c is observed upon cooling (cf. inset of Fig. 1) indicating strong deviations from LFL behavior as expected for systems close to a QCP. Below 1 K the data can be fit by $\alpha(T)/T = aT^{-0.5} + b$, i.e., the sum of a singular and a normal contribution. Such a temperature dependence, also seen for $CeNi_2Ge_2$ [21], has been predicted very recently by the itinerant spin-density-wave (SDW) theory for three-dimensional (3D) critical spin fluctuations at an AF QCP [22].

Now we turn to the pronounced anisotropy of the normal-state thermal expansion at $B = 0$. Provided the low-T thermodynamic properties are governed by a single characteristic energy scale, $k_B T^*$, the experimentally derived Grüneisen parameter measures the volume dependence of this characteristic energy: $\Gamma = -\partial \ln T^*/\partial \ln V$. Likewise, for anisotropic systems the uniaxial Grüneisen parameters Γ_i give the uniaxial-strain dependences of T^*. For tetragonal systems, the values for $\Gamma_{a,c}$ are obtained by calculating $\Gamma_a = V_{mol}[2c_{13}\alpha_a + c_{33}\alpha_c]/C$ and $\Gamma_c = V_{mol}[(c_{11} + c_{12})\alpha_a + c_{13}\alpha_c]/C$ [23] using the specific heat reported in [13] and the elastic constants c_{ij} measured for $CeRhIn_5$ [24] (no significant differences are expected for c_{ij} within the $CeMIn_5$ series). For $CeIrIn_5$, Γ_c is found to be about 2.5× larger than Γ_a at T_c (Fig. 2). Thus the characteristic Kondo-lattice energy $k_B T^*$ is more sensitive to strain along the c axis compared to in-plane strain. This observation is counterintuitive to viewing $CeIrIn_5$ a quasi-two-dimensional HF system for which the in-plane strain effects associated with changes of the Ce-In(1) hybridization should predominate those along the c axis: Our results clearly demonstrate that the Ce-In(2) hybridization is more important for the low-T properties.

$CeCoIn_5$ has the record T_c among the Ce- and U-based HF superconductors. Like $CeIrIn_5$ this system, too, shows an anisotropic normal-state thermal expansion (Fig. 3). The uniaxial pressure dependences of T_c calculated from

TABLE I. Values for the SC transition temperature T_c (at $B = 0$), molar volume V_{mol}, specific heat jump at T_c, $\Delta C/T$, taken from [13] ($CeIrIn_5$) and [14] ($CeCoIn_5$), uniaxial pressure dependences of T_c in the limit of vanishing pressure, $\partial T_c/\partial p_c$ and $\partial T_c/\partial p_a$, calculated hydrostatic pressure dependence, $\partial T_c/\partial p_V = 2\partial T_c/\partial p_a + \partial T_c/\partial p_c$, and the hydrostatic pressure dependence, $\partial T_c/\partial p_{hydr}$ measured by specific heat on both $CeIrIn_5$ [18][a], and $CeCoIn_5$ [19][b], and by resistivity on $CeCoIn_5$ [20] [c].

	T_c (K)	V_{mol} (m^3/mol)	$\Delta C/T$ (J/mol K^2)	$\partial T_c/\partial p_c$ (mK/GPa)	$\partial T_c/\partial p_a$ (mK/GPa)	$\partial T_c/\partial p_V$ (mK/GPa)	$\partial T_c/\partial p_{hydr}$ (mK/GPa)
$CeIrIn_5$	0.38	9.86×10^{-5}	0.55	-890 ± 40	$+540 \pm 40$	$+190 \pm 60$	+250[a]
$CeCoIn_5$	2.23	9.72×10^{-5}	1.3	$+75 \pm 10$	$+290 \pm 30$	$+655 \pm 50$	+400[b], +500[c]

[a]Measured by specific heat on $CeIrIn_5$ [18].
[b]Measured by specific heat on $CeCoIn_5$ [19].
[c]Measured by resistivity on $CeCoIn_5$ [20].

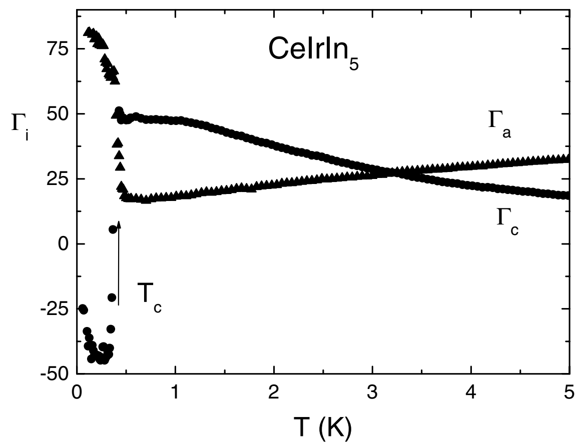

FIG. 2. Uniaxial Grüneisen parameters, Γ_a and Γ_c, as defined in the text, vs T for CeIrIn$_5$.

the $\alpha(T)$ discontinuities at T_c reveal a small positive value for uniaxial pressure applied parallel to the c axis and a much larger positive value for uniaxial pressure applied along the a axis (see Table I). Thus, in contrast to CeIrIn$_5$ a reduction of the lattice parameter along both directions yields an increase of T_c in CeCoIn$_5$. Again, the calculated hydrostatic pressure dependence is in good agreement with the results obtained via specific heat [19] and resistivity [20] measurements under hydrostatic pressure. The upper critical field $B_{c2}(T)$ shows a pronounced anisotropy with $B_{c2}(0)$ values of about 4.9 T for $B \parallel c$ and 11.8 T for

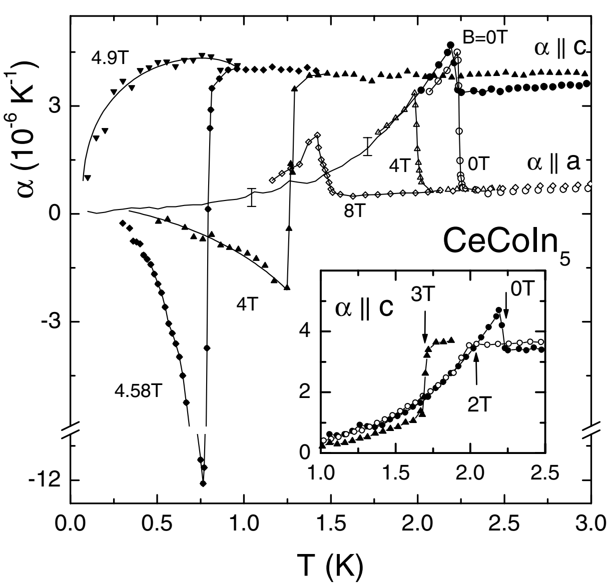

FIG. 3. Linear thermal expansion coefficients, α_a (open symbols) and α_c (solid symbols), vs T for CeCoIn$_5$ at differing magnetic fields. The line shows the curve for $\alpha_c(B = 0)$. Within the error bar the solid line represents also the α_c data at 0 T, 4 T, and 8 T in the SC state. The inset enlarges $\alpha_c(T)$ at low fields. Arrows denote superconducting transitions.

$B \parallel a$ [14]. For $B \parallel c$ we observe a quite unusual behavior in thermal expansion: The small positive jump in $\alpha_c(T)$ observed at $B = 0$ is suppressed with increasing B and vanishes completely for $B = 2$ T (cf. inset of Fig. 3). For larger B fields, an increasingly large negative jump anomaly develops. Furthermore, an unexpected sharpening of the transition occurs for $B \geq 4$ T. At $B = 4.58$ T we observe $\alpha_c(T)$ to almost diverge at T_c. This corroborates specific-heat results [25] indicating that the superconducting transition becomes of first order below $T \approx 0.7$ K and above $B \approx 4.6$ T. In this parameter range our magnetostriction experiments reveal a steplike change upon crossing the $B_{c2}(T)$ boundary [25]. This anomaly which can be related to Pauli limiting as a consequence of the large spin susceptibility for $B \parallel c$ is discussed elsewhere [25].

The normal-state specific heat coefficient, $C(T)/T$, measured at $B = 5\,\mathrm{T} \gtrsim B_{c2}(0)$ ($B \parallel c$) was found to follow a $\log(T_0/T)$ dependence in a wide temperature window, $0.3\ \mathrm{K} \leq T \leq 8\ \mathrm{K}$ [26]. As shown in the upper part of Fig. 4, the thermal expansion coefficient $\alpha_c(T)$ measured under the same conditions reveals an even stronger divergence, i.e., $\alpha_c/T \propto a_1 + a_2/T$. Thus, also the Grüneisen ratio being proportional to α/C diverges to the lowest temperatures, which provides clear-cut evidence for CeCoIn$_5$ at $B = B_{c2}(0)$ being located near a magnetic QCP [22]. A very similar conclusion has been drawn very recently from electrical resistivity measurements that revealed a $1/(B - B_c)^{1.37}$ divergence of the coefficient $A(B)$ of the field induced $\rho - \rho_0 = A(B)T^2$ behavior for $B > B_{c2}(0)$ [27]. Most remarkably, the divergence of the normal-state α_c/T is stronger than expected by the 3D SDW theory, but weaker than the $a_1T^{-1}\log[b\log(T_0/T)] + a_2$ form expected in a 2D SDW picture [22]. This raises the question of whether

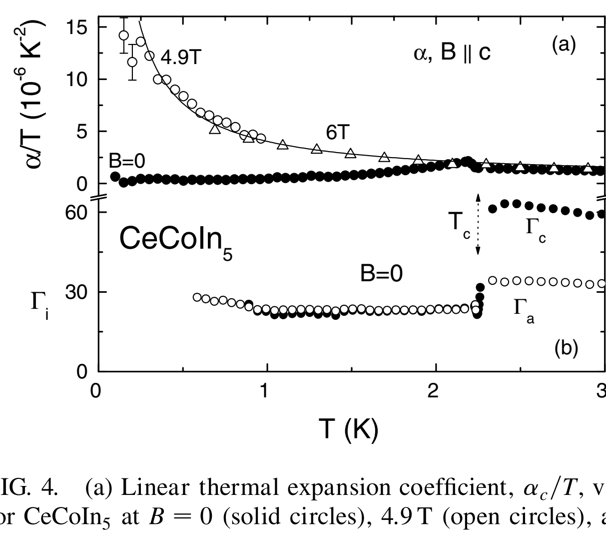

FIG. 4. (a) Linear thermal expansion coefficient, α_c/T, vs T for CeCoIn$_5$ at $B = 0$ (solid circles), 4.9 T (open circles), and 6 T (open triangle). The line represents $\alpha/T = a_0 + a_1/T$ with $a_0 = 0.40 \times 10^{-6}\,\mathrm{K}^{-2}$ and $a_1 = 3.64 \times 10^{-6}\,\mathrm{K}^{-1}$. (b) Uniaxial Grüneisen parameters, Γ_a and Γ_c, vs T for CeCoIn$_5$ at $B = 0$.

the itinerant SDW theory is applicable to $CeCoIn_5$ [28]. At $B = 0$, i.e., in the SC state, the Grüneisen parameter is completely isotropic indicating that the SC gap depends isotropically upon uniaxial lattice distortions, strikingly different to the case of $CeIrIn_5$ (cf. Fig. 2).

The pronounced deviations from LFL behavior observed at overcritical magnetic fields down to lowest T in $CeIrIn_5$ and $CeCoIn_5$, along with the positive $\partial T_c/\partial p_{\mathrm{hydr}}$ values, show clearly that both compounds are located close to the magnetic instability at the left side of the superconducting "dome" found in the T_c vs p phase diagram [29]. As reported for other HF superconductors [30,31], superconductivity develops out of a non-Fermi-liquid state. By comparing the SC phase-transition anomalies observed in $\alpha(T)$ for $CeIrIn_5$ and $CeCoIn_5$, we emphasize that at least two parameters determine the strength of the SC pairing interaction in $CeIrIn_5$ and $CeCoIn_5$. The first one is the crystalline anisotropy as indicated by the opposite signs of the uniaxial pressure dependences of T_c for $CeIrIn_5$. For a series of $CeM_{1-x}N_xIn_5$ ($M, N =$ Ir, Co, Rh) compounds a correlation between the c/a ratio and T_c was observed [32]. The positive value of $\Delta\alpha_c(B = 0)$ for $CeCoIn_5$ is, however, in contradiction to this simple relation and, furthermore, highlights the importance of a second parameter that is measuring the strength of the $4f$-conduction electron hybridization. The latter is most sensitive to strain along the c axis, as indicated by the roughly $2\times$ higher Γ_c values at T_c compared to Γ_a observed for both systems. This observation is incompatible with the simple view of a quasi-two-dimensional system suggested by the partly cylindric Fermi surface [33,34]. Thus, uniaxial pressure along the c axis has a twofold effect: It increases the hybridization strength most effectively, although it reduces the crystalline anisotropy, and leads finally to a rise of T_c. The expected negative uniaxial pressure dependence of T_c for pressure along the c axis is observed for magnetic fields larger than 2 T, as is evident from the unusual sign change of $\Delta\alpha_a$ (cf. inset of Fig. 3).

To summarize, the high T_c values observed in the $CeMIn_5$ family compared to other Ce-based HF superconductors are most likely caused by their crystalline anisotropy. Furthermore, they seem to be closely related to critical fluctuations near a magnetic instability, in qualitative agreement with the predictions for spin-fluctuation mediated superconductivity near a quantum-critical point [10]. Remarkably, the characteristic energy scale of the $CeMIn_5$ systems depends much more strongly on changes of the c-axis compared to the a-axis lattice parameter.

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