Towards the Identification of a Quantum Critical Line in the (p, B) Phase Diagram of $CeCoIn_{5}$ with Thermal-Expansion Measurements

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Towards the identification of a quantum critical line in the (p, B) phase diagram of $CeCoIn_5$ with thermal-expansion measurements

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The low-temperature thermal expansion of $CeCoIn_5$ single crystals measured parallel and perpendicular to magnetic fields B oriented along the c axis yields the volume thermal-expansion coefficient β. Considerable deviations of $\beta(T)$ from Fermi-liquid behavior occur already within the superconducting region of the (B,T) phase diagram and become maximal at the upper critical field B^0_{c2}. However, $\beta(T)$ and the Grüneisen parameter Γ are incompatible with a quantum critical point (QCP) at B^0_{c2}, but allow for a QCP shielded by superconductivity and extending to negative pressures for $B < B^0_{c2}$. We construct a tentative (p, B, T) phase diagram of $CeCoIn_5$ suggesting a quantum critical line in the (p,B) plane.

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Many Ce-based heavy-fermion compounds exhibit unconventional superconductivity on the verge of long-range magnetic order, i.e. in the vicinity of magnetic quantum critical points (QCPs) [1, 2]. The presence of these magnetic instabilities is manifested by profound deviations from Fermi-liquid (FL) behavior [3]. $CeCoIn_5$ is a typical representative of these systems with pronounced non-Fermi-liquid (NFL) behavior at temperatures above its superconducting transition at $T_c = 2.3\,\mathrm{K}$ [4]. With increasing magnetic field B the deviations from FL behavior become maximal at the upper critical field $B^0_{c2} = B_{c2}(T \to 0) \approx 5\,\mathrm{T}$ for $B \parallel c$ suggesting that NFL behavior might originate from a nearby field-induced antiferromagnetic (AF) QCP at B^0_{c2}. Indeed, the low-temperature dependence of the specific-heat coefficient C/T at B^0_{c2} follows the Hertz-Millis-Moriya (HMM) theory [5–7] of a two-dimensional spin-density-wave instability, as it might be expected for the layered, tetragonal crystal structure of $CeCoIn_5$ [4, 8]. The linear thermal-expansion coefficient along the c axis α_c [9], the resistivity, and the thermal conductivity [10], however, reveal at $T \leq 0.3\,\mathrm{K}$ deviations from HMM behavior that were attributed to a dimensional crossover for $T \to 0$ [9]. Hall-effect measurements indicate a QCP at the second-order phase boundary of the recently discovered Q phase [11, 12], possibly indicating that critical fluctuations of the Q phase produce NFL behavior. In contrast, resistivity measurements point to a QCP at negative hydrostatic pressures p [13] or emerging from a pressure- and field-dependent critical line that is masked by superconductivity [14].

To search for the origin of NFL behavior of $CeCoIn_5$ in the (p,B) plane, we have exploited a characteristic QCP feature: the accumulation of entropy at finite temperatures due to the instability of the ground state [15]. It can be accessed from the derivatives $\partial S/\partial p$, or $\partial S/\partial B$, the volume thermal-expansion coefficient $\beta = -V^{-1} \cdot \partial S/\partial p$ (V = molar volume) and the magnetization $\partial M/\partial T = \partial S/\partial B$, respectively. Measurements of M point to quantum criticality directly at or below B^0_{c2} [16].

To study $\partial S/\partial p$, we performed thermal-expansion measurements in magnetic fields of up to $B = 14\,\mathrm{T}$ with $B \parallel c$ between 40 mK and 4 K. As an important extension to previous measurements [9, 17] our capacitive dilatometer allows for thermal expansion and magnetostriction measurements both parallel and perpendicular to the applied magnetic field with a resolution of $\Delta l = 10^{-3}\,\mathrm{\AA}$. $CeCoIn_5$ single crystals were grown in In flux. The plate-like crystals have typical dimensions of $l_a \approx 3\,\mathrm{mm}$ and $l_c \leq 0.5\,\mathrm{mm}$ thus enabling a relative resolution of $\Delta l/l \approx 10^{-9}$. To obtain the Grüneisen parameter Γ at high fields, we measured the specific heat $C(T)$ between 350 mK and 5 K at $B = 10\,\mathrm{T}$ and 14 T with a Physical Properties Measurement System from Quantum Design.

In a Fermi liquid the thermal-expansion coefficient approaches a linear temperature dependence for $T \to 0$, proportional to the specific heat. The proportionality constant is essentially the Grüneisen parameter $\Gamma = (V/\kappa_s) \cdot \beta/C$ (κ_s = adiabatic compressibility) and reflects the volume dependence of the Fermi energy. To expose deviations from FL behavior in $CeCoIn_5$, we plot the volume thermal-expansion coefficient, calculated from the measured linear thermal-expansion coefficients $\beta = 2\alpha_a + \alpha_c$, as β/T in Fig. 1(a) and (b) for fields below and above B^0_{c2}, respectively. In zero magnetic field, β/T drops asymptotically towards zero for decreasing T, just as C/T, due to the presence of the superconducting gap in the energy spectrum of the quasiparticles. At higher fields, still in the field range of the second-order superconducting phase transition at B_{c2}, β/T grows and starts to diverge for $T \to 0$. Usually, in the vortex phase of a conventional type-II superconductor β/T reaches a finite constant low-temperature value due to the remanent Fermi liquid in the vortex cores. Although the vortex lattice in $CeCoIn_5$ is less dense due to the Pauli-limited upper critical field the discussion in the following sections will show that this upturn is not related to the pressure and temperature dependence of B_{c2}. Another possible explanation is given by a non-Fermi liquid behavior of the normal conducting vortex cores. This seems to be supported by specific-heat measurements where the entropy conservation requires a continuously increasing $C/T \propto -\log T$ of the normal conducting background for $T \rightarrow 0$ [3, 4].

β/T exhibits the strongest low-temperatures upturn close to B^0_{c2} [Fig. 1(a)]. Further enhanced fields weaken the divergence and lead, in a growing temperature range at $T < T_{\rm FL}$, to the restoration of the Fermi liquid, i.e. β/T = const [Fig. 1(b)]. This clear feature does not support an interpretation [9] in terms of a dimensional crossover. We emphasize that at low temperatures β/T remains positive in the entire measured field range, except for the peaked anomaly of the first-order transition [Fig. 1(c)]. This is an important result, because the absence of a sign change demonstrates that S has no maximum as a function of p at B^0_{c2} but grows further under negative, hydrostatic pressure or, equivalently, with increasing unit-cell volume. These results speak against a QCP in the vicinity of $B^0_{c2}(p = 0)$.

A direct proof for the existence of a pressure-tuned QCP is the Grüneisen parameter Γ. If a system is dominated by a single characteristic energy scale $E^*(V)$, Γ reflects its volume dependence: $\Gamma = -\partial \ln E^*/\partial \ln V$. Since at a QCP E^* vanishes, Γ is expected to diverge when the QCP is approached [18]. We estimate Γ by assuming κ_s to be roughly equal to the isothermal compressibility of CeCoIn_5 $\kappa_T = 1.31 \cdot 10^{-2}\,\text{GPa}^{-1}$ [19] and with specific-heat data with the hyperfine contribution subtracted from [8, 20, 21] and our measurements. The resulting $\Gamma(T)$ curves, shown in Fig. 2(a), exhibit high $\Gamma(T)$ values up to 80, typical for heavy-fermion systems [22], that illustrate their high sensitivity to volume changes. The $\Gamma(T)$ curve at $B = 5.1\,\text{T} \approx B^0_{c2}$ is the highest of all fields. Here, Γ initially shows a rise with decreasing T, but levels off below $\approx 260\,\text{mK}$ and drops again. Although the strongly enhanced Γ points to a pressure-tuned QCP, the lack of a low-temperature divergence provides clear evidence against a QCP at ambient pressure in the entire field range up to 14 T.

To clarify a possible link between superconductivity and quantum criticality in CeCoIn_5, we compare the volume dependence of T_c with the measured Grüneisen parameter. If superconductivity provides the dominant energy scale $E^* \propto T_c$, Γ is determined by $-d\ln T_c/d\ln V = (T_c\kappa_s)^{-1} \cdot dT_c/dp$ at $T << T_c$. The hydrostatic pressure dependence of the second order transition $dT_c/dp = 2dT_c/d\sigma_a + 2dT_c/d\sigma_c$ is calculated from the uniaxial pressure (σ_i) dependences using the Ehrenfest relation $dT_c/d\sigma_i = T_cV \cdot \Delta\alpha_i/\Delta C$ ($i = a, c$). Here, $\Delta\alpha_i$ and ΔC are the discontinuities of the linear thermal-expansion coefficients and the specific heat at T_c, respectively. The resulting dT_c/dp data are displayed as a function of B in Fig. 2(b). They are in good agreement with experiments under hydrostatic pressure [23]. dT_c/dp shows a sign change at $B_+ \approx 4.1 \pm 0.2\,\text{T}$. At higher fields, when the transition becomes first-order, dT_c/dp remains negative up to B^0_{c2} as demonstrated by the negative volume discontinuities at T_c [Fig. 1(a)], which is expected to produce a likewise negative low-temperature Grüneisen pa-

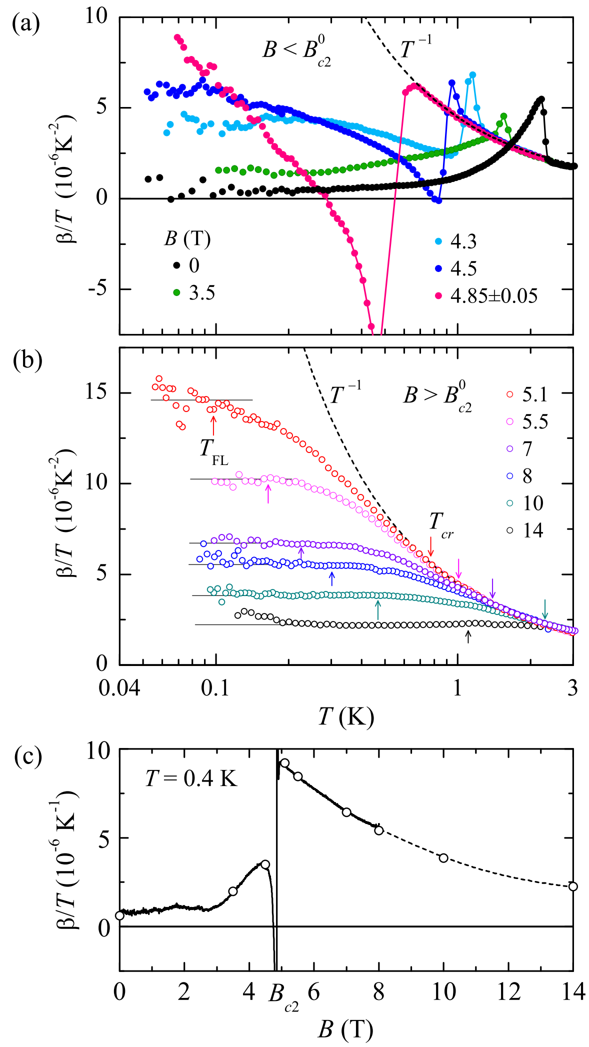

FIG. 1. (Color online) The volume thermal-expansion coefficient of CeCoIn_5 divided by temperature β/T in magnetic fields parallel to the c axis below (a) and above B^0_{c2} (b). In order to optimize the resolution of the experiment, different samples were used to measure α_a and α_c. They have slightly different B^0_{c2} values of $\approx 5.1\,\text{T}$ and $\approx 5.05\,\text{T}$, respectively. Therefore, the data for $B = 4.85\,\text{T}$ are a combination of measurements at $B = 4.8\,\text{T}$ and $4.9\,\text{T}$ where the T_c values are identical. The dashed lines represent the non-Fermi-liquid behavior for the HMM model. (c) β/T as a function of B at $T = 0.4\,\text{K}$ obtained from magnetostriction measurements (continuous line) and from temperature-dependent measurements at constant B (circles). Dashed line is a guide to the eye.

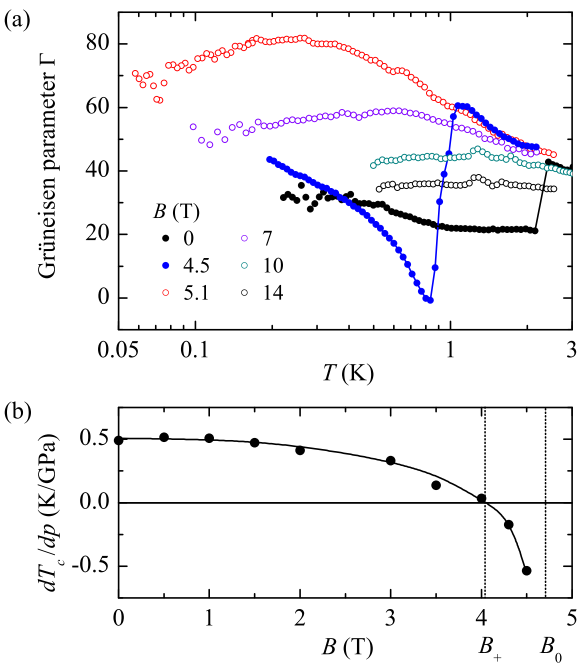

FIG. 2. (Color online) (a) The Grüneisen parameter Γ versus T at different constant fields parallel to the c axis. (b) The hydrostatic pressure dependences dT_c/dp as a function of B. Lines are guides to the eye.

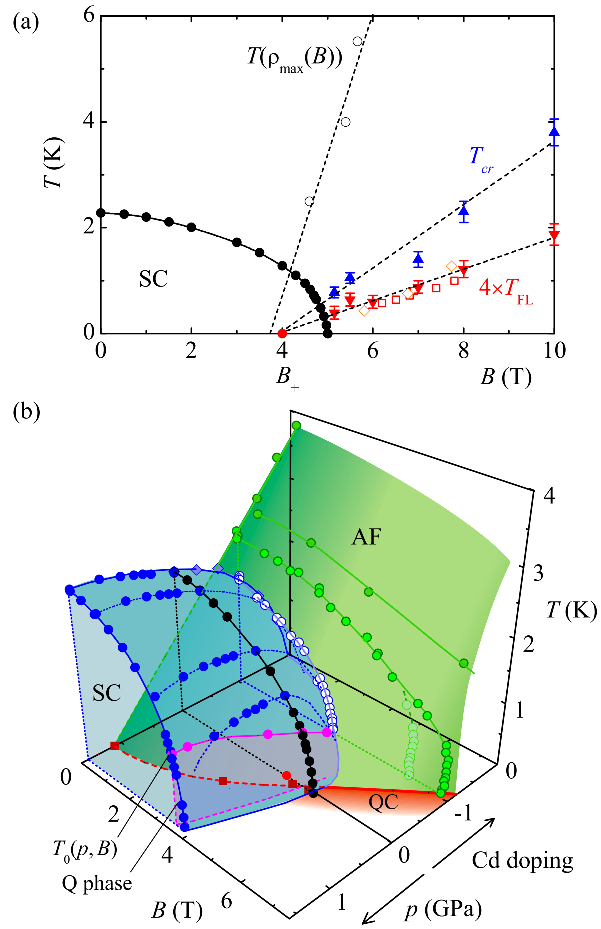

FIG. 3. (Color online) (a) The (B, T) phase diagram of $CeCoIn_5$ at ambient pressure constructed from our measurements of the superconducting transition (●), the onset of FL behavior at T_{FL} (▲), and the change in the critical behavior at T_{cr} (▼). In addition, literature data are shown for the maximum of the magnetoresistance $T(\rho_{max}(B))$ (○ [24]) and T_{FL} determined by Hall effect (□ [25]) and resistivity measurements (◇ [26]). (b) Tentative (p, B, T) phase diagram of $CeCoIn_5$. The data from (a) have been extended by published measurements under hydrostatic (● [14, 27–29]) and under negative, chemical pressure on $CeCoIn_{5-x}Cd_x$ (●,○ [30–32]). To account for the different sample qualities and varying T_c definitions the measurements have been scaled to match in the regions of overlap. B_+ (●), and the onset of the NFL behavior (■ [14]) are projected onto the $T = 0$ plane.

rameter in the field range $B_+ < B < B^0_{c2}$. Experimentally, however $\Gamma(T)$ exhibits large positive values in the entire field range for $T \to 0$, apart from the small field internal of the first-order transition near B_{c2} [Fig. 1(c)]. Therefore critical fluctuations of superconductivity or the first-order transition of the Q phase reflecting the temperature and pressure dependence of the Pauli-limited B_{c2} cannot be responsible for the NFL behavior visible in the low-temperature upturn of β/T.

The field dependence of the FL recovery [Fig. 1(b)] can be used to estimate a possible field-induced origin of the NFL behavior in $CeCoIn_5$ at $p = 0$. To look for such a behavior with a decrease of T_{FL} toward a QCP, we construct from our data the (B, T) phase diagram in Fig. 3(a). Within the experimental error, T_{FL} agrees with the onset of the T^2 dependence of the resistivity ρ [10, 26] or of constant C/T values [8]. Other characteristic features that are related to the FL recovery such as the minimum of the differential Hall coefficient [25], coincide likewise with T_{FL}. A striking feature of the phase diagram is that T_{FL} does not vanish at B^0_{c2}. For $B \leq 10\,\mathrm{T}$, T_{FL} extrapolates linearly to a $T = 0$ critical field of $B_c = 4.1 \pm 0.1\,\mathrm{T}$, which is equal to B_+ where, according to $dT_c/dp = 0$, T_c exhibits a maximum as a function of p. This suggests that the (hidden) AF QCP at $p = 0$ is expected to occur at the field where $T_c(p)$ of the superconducting dome is maximal. Furthermore, the evolution of the coherence maximum of the magnetoresistance $\rho(B)$ with temperature, $T(\rho_{max}(B))$, [Fig. 3(a)] [24] extrapolates to roughly the same B_c for $T \to 0$. Therefore a QCP at B_c has been postulated [25]. In the $T = 0$ plane with two control parameters p and B, a line of QCPs is highly plausible. This is supported by the thermal-expansion measurements above T_c [Fig. 1(a)] or above T_{FL} [Fig. 1(b)] where all β/T curves merge into the same critical T^{-1} dependence. Therefore, the NFL behavior at different fields is likely to have a common origin. The NFL behavior might be explained by a single QCP at negative critical pressure p_c and $B \approx B_+$ or, alternatively by an extended quantum critical phase boundary. Indeed, high-pressure experiments indicate that the NFL behavior emanates from a p- and B-dependent line, which is shielded by superconductivity [14].

To check whether a quantum critical line is compatible with the p dependence of T_c and the NFL behavior from our measurements, we extend the phase diagram to finite pressures with literature data measured under hydrostatic pressure or on Cd-doped $CeCoIn_5$ [Fig. 3(b)]. Although Cd doping does not generate a noteworthy enlargement of the unit cell, pressure experiments on $CeCoIn_{5-x}Cd_x$ show that a nominal Cd concentration of 5% correponds to a negative chemical pressure of -0.7 GPa [33]. Doping with Cd suppresses superconductivity and leads to long-range AF order. At $B = 0$, a linear extrapolation of the Néel temperature T_N to positive pressures reveals that, without intervening superconductivity, AF order would disappear at a pressure where T_c becomes maximal as indeed shown above for $B = B_c \approx B_+$. This is a common feature of many unconventional superconductors close to AF order and is generally taken as evidence for superconductivity mediated by magnetic fluctuations [1, 2]. As shown in the $T = 0$ plane of Fig. 3(b), with increasing B, the T_c maximum follows approximately the origin of NFL behavior to lower pressures until it reaches at $p = 0$ a field that is close to B_+ (or B_c). At higher fields, especially at B^0_{c2}, the phase line continues to negative pressures, again in accordance to our findings. In contrast to the p and B dependence of the NFL behavior, the onset of the discontinuous superconducting transition and Q phase $B_0(p)$ follows the upper critical field $B^0_{c2}(p)$ and is still present when, beyond $p > p_c$, NFL behavior can no longer be observed. Although, at ambient pressure, B_0 and B_+ are close to each other, their qualitatively different pressure dependence gives clear evidence that the Q phase can be ruled out as source for the NFL behavior. The close relationship between the T_c maximum, the origin of NFL behavior, and the endpoint of AF order, on the other hand, suggest that the quantum critical line corresponds to the AF phase boundary. In this case, the available data can be described by the tentative phase diagram proposed in Fig. 3(b).

In summary. we performed thermal-expansion measurements at different magnetic fields to study the pressure and field dependence of the non-Fermi-liquid behavior in $CeCoIn_5$ which extends at ambient pressure over a wide field range. Although the deviations from Fermi-liquid behavior reach a maximum at the onset of superconductivity at B^0_{c2}, the Grüneisen parameter clearly demonstrates that at $p = 0$ no QCP exists up to a field of 14 T. Due to the finite distance of the QCP from B^0_{c2}, the critical behavior changes at low temperatures toward Fermi-liquid recovery. A combination of our results with literature data allows to construct a tentative (p, B, T) phase diagram, in which the NFL behavior originates from a quantum critical line of the onset of antiferromagnetic order. This line, emanating from a point of positive pressure on the $B = 0$ axis, is hidden by the superconducting dome. It may extend to negative pressures at magnetic fields larger than B^0_{c2} as suggested by recent experiments on Cd-doped $CeCoIn_5$.

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