Zero-Field Quantum Critical Point in $CeCoIn_5$

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Quantum criticality in the normal and superconducting states of the heavy-fermion metal $CeCoIn_5$ is studied by measurements of the magnetic Grüneisen ratio Γ_H and specific heat in different field orientations and temperatures down to 50 mK. A universal temperature over magnetic field scaling of Γ_H in the normal state indicates a hidden quantum critical point at zero field. Within the superconducting state, the quasiparticle entropy at constant temperature increases upon reducing the field towards zero, providing additional evidence for zero-field quantum criticality.

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The interplay between magnetism and unconventional superconductivity is one of the central issues in condensed matter physics. Several material classes such as cuprates, iron pnictides, or heavy-fermion metals display non-Fermi-liquid (NFL) normal-state properties that may arise from a quantum critical point (QCP) at which a long-range ordered phase is continuously suppressed to zero temperature [1–3]. However, NFL properties in the vicinity of such hidden QCPs cannot be investigated without destroying the superconducting (SC) state by sufficiently large magnetic fields, which also strongly influence quantum criticality [4,5]. On the other hand, if there is universal scaling of the free energy with respect to temperature and some control parameter in the normal state, it is possible to prove the existence and characterize the nature of the hidden QCP. Most sensitive probes of such scaling behaviors are the Grüneisen ratios $\Gamma_r = T^{-1}(dT/dr)_S$ (r, pressure or magnetic field; S, entropy), which diverge in the approach of the QCP [6]. Since magnetic field can easily be varied *in situ* and the magnetic Grüneisen ratio which equals the adiabatic magnetocaloric effect divided by temperature is directly measurable with high precision, a field-tuned QCP hidden by superconductivity as proposed for the heavy-fermion metal $CeCoIn_5$ [7–12] or Ce_2PdIn_8 [13,14] should best be characterized by the latter property.

$CeCoIn_5$ undergoes a SC transition at $T_c = 2.3$ K, which is the highest among the ambient-pressure heavy-fermion superconductors [15] but low enough to neglect phononic contributions to heat capacity and suppress the SC state by moderate magnetic fields. $CeCoIn_5$ has attracted considerable attention as very clean metal close to long-range magnetic order, which displays intriguing SC and normal state properties [7–12,16,17]. Electrical resistivity, specific heat, and thermal expansion display NFL behavior in the normal state above T_c which extends to mK temperatures close to the SC upper critical field H_{c2} [7–12,18]. On the other hand, for fields sufficiently larger than H_{c2}, a crossover to Fermi-liquid (FL) behavior has been recovered, which allows us to extrapolate to a field-induced QCP. Early electrical resistivity [7] and specific heat [8] measurements suggest a field-induced QCP very close to the upper critical field, which amounts to $H_{c2} = 5$ and 12 T for the field along and perpendicular to [001], respectively. Subsequent Hall effect [19] and thermal expansion [11] measurements, however, have extrapolated the critical field H_c for the field-tuned QCP to values clearly below H_{c2}, i.e., around 4 T for $H \parallel [001]$. A peculiar observation is the dependence of estimated H_c on the current direction of electrical resistivity for $H \parallel [001]$. The A coefficient of FL resistivity $\rho = \rho_0 + AT^2$, measured with the current along the basal plane, diverges towards 5 T [7,20,21], while A with the current along [001] indicates a significantly lower field for the singularity 1.5–3 T [22]. In view of the controversy concerning the exact location of a possible field-induced QCP in $CeCoIn_5$, systematic studies of the magnetocaloric effect, which is the most sensitive thermodynamic probe of field-tuned quantum criticality, are highly desirable.

Below, we report a systematic investigation of the temperature, field, and field-angle dependence of the magnetic Grüneisen parameter in the normal state of $CeCoIn_5$. Surprisingly, we have discovered universal quantum critical scaling, indicating a zero-field QCP. This is further supported by an enhanced quasiparticle entropy, derived from the magnetic Grüneisen ratio and specific heat within the SC state in the vicinity of zero field. This implies that $CeCoIn_5$ is exceptional as clean material at a QCP without additional fine-tuning of composition, pressure, or magnetic field.

High quality single crystals were grown by the self-flux method. The specific heat $C(T, H)$ and magnetic Grüneisen ratio $\Gamma_H = T^{-1}(dT/dH)_S$ were measured with very high resolution in a dilution refrigerator with a SC magnet equipped with an additional modulation coil by utilizing heat-pulse and alternating field techniques, respectively, as described in Ref. [23]. The magnetic field has been applied along four different field angles ranging from the [001] to the [100] direction.

We first focus on the magnetic Grüneisen ratio in the normal state at various fields and field orientations, cf. Fig. 1. Upon cooling from high temperatures, $\Gamma_H(T)/H$ first increases until it passes a maximum and, as most clearly seen for fields above 6 T, saturates at lowest temperatures. Such temperature dependence is characteristic for the crossover between NFL behavior at high T and a FL state at low T [24], which, e.g., at 5 T, occurs near 0.14 K. The data are thus incompatible with a QCP at H_{c2}. As shown in Fig. 1(b), similar behavior is also found for $H \parallel [100]$ and all intermediate field orientations. Within the quantum critical regime, $\Gamma_H(T)$ is expected to display a power-law divergence upon cooling. However, for fields $H > H_{c2}$, we only observe an almost linear increase on the semilog scale upon cooling. This indicates

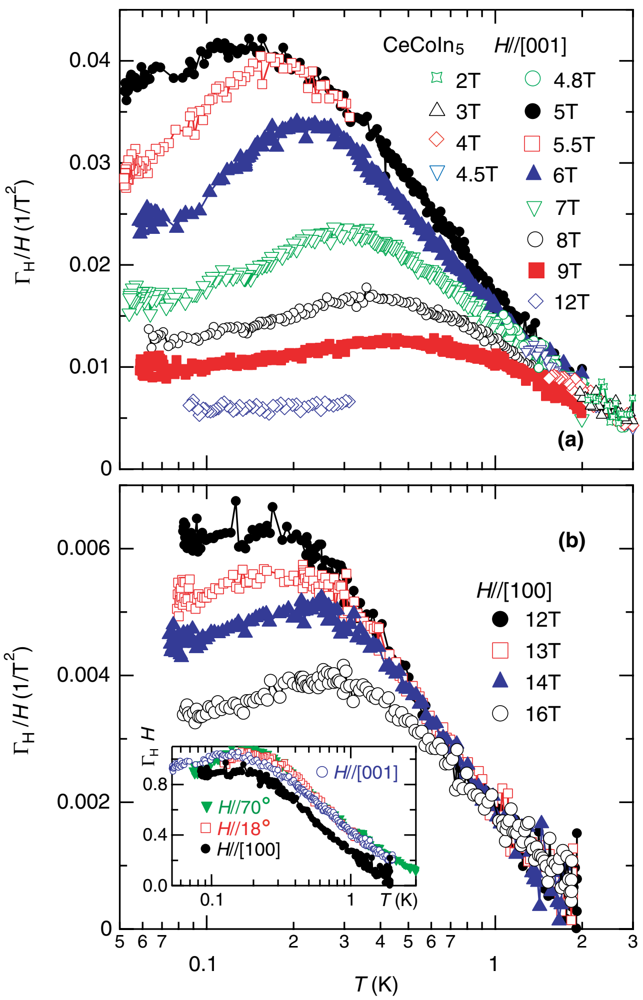

FIG. 1 (color online). Magnetic Grüneisen ratio divided by magnetic field Γ_H/H of $CeCoIn_5$ in the normal conducting state plotted against temperature for fields applied along (a) [001] and (b) [100]. The inset displays $\Gamma_H H$ as a function of temperature for different field angles close to the respective upper critical fields. Labels 18° and 70° denote field angles from [100] towards [001]. The applied magnetic field is 5, 6, 10, and 12 T for $H \parallel [001]$, 70°, 18°, and parallel [100], respectively.

that the QCP must be far below H_{c2}. Furthermore, Γ_H must change its sign across a QCP [24], while the measured normal-state magnetic Grüneisen ratio is always positive even below 2 T.

Further information on the critical field H_c of the QCP can be obtained by analyzing the magnetic Grüneisen ratio in the FL state for $T \to 0$. For a field-induced QCP which follows universal scaling, it is expected that $\Gamma_H(H - H_c) = -\nu(d - z)$, where d is the spatial dimension, ν the correlation length exponent, and z the dynamical critical exponent [6,24]. Thus, if the data follow such behavior, we may determine H_c and obtain important information on the quantum critical exponents. For the analysis, we include data for fields parallel and perpendicular to the [001] direction, as well as for two different intermediate field directions at the respective upper critical fields [cf. inset of Fig. 1(b)]. The overall temperature dependencies for all these field directions are similar, and $\Gamma_H H$ approaches a common value for $T \to 0$. As shown below, this is a consequence of universal quantum critical scaling, since the above prefactor $-\nu(d - z)$ characterizes the nature of quantum criticality and is independent of the direction of applied field. Furthermore, it indicates a quantum critical field H_c being close to zero.

In inset (a) of Fig. 2, we plot the inverse of Γ_H for $T \to 0$ in the FL regime at various different field values. Note that

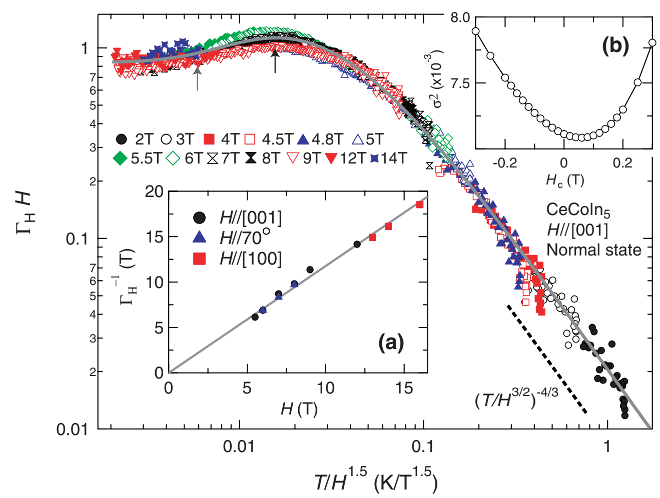

FIG. 2 (color online). Magnetic Grüneisen ratio of $CeCoIn_5$ for the field along [001] as $\Gamma_H H$ versus $T/H^{3/2}$, on double logarithmic scale. The solid grey line indicates a phenomenological fit by the function $f(x)$ (see the main text for the definition). The positions of the maximum and inflection points, respectively, 0.015 and 0.006 $\mathrm{K\,T^{-3/2}}$, that define the crossover between the NFL and FL regimes, are indicated by arrows. Inset (a) shows Γ_H^{-1} within the FL regime for $T \to 0$ versus magnetic field for different field directions. The solid grey line indicates a divergence of the magnetic Grüneisen ratio as $\Gamma_H = 0.85/H$. Inset (b) shows the weighted mean-square deviation from the phenomenological function $f(x)$ versus the quantum critical field H_c.

an intercept of $\Gamma_H^{-1} = 0$ corresponds to the quantum critical field H_c, analogous to the Weiss temperature in a Curie-Weiss plot. Remarkably, all data points collapse on a universal line through the origin, regardless of the field direction. Given the sizable magnetic anisotropy of this system [25], this isotropic divergence of the magnetic Grüneisen ratio towards zero field with a common prefactor provides strong evidence for universal quantum critical scaling with H_c close to zero.

Following the theory of Refs. [6,24,26], we assume that critical behavior is governed by a single diverging time scale near the QCP. The critical contribution to the free energy $F_{\rm cr}$ for magnetic field as a tuning parameter can then be expressed by

$$F_{\rm cr} = aT^{(d+z)/z}\phi\left(\frac{bh}{T^{1/\nu z}}\right), \tag{1}$$

where $h = H - H_c$, and a and b are nonuniversal constants. The thermodynamic properties are then expressed by derivative(s) of the free energy and therefore should collapse on a scaling function of $T/h^{\nu z}$. All our Γ_H data collapse on a single curve in a log-log scaling plot of the form $\Gamma_H H$ versus $T/H^{3/2}$ (Fig. 2), indicating that $h = H$ equivalent to $H_c = 0$ and implying $\nu z = 3/2$. Within the FL regime at low temperatures and $H > 0$, the data scatter around a constant value of $\Gamma_H H \approx 0.85$ (consistent with the slope of the line in the inset of Fig. 2), whose meaning is addressed below. We first focus on the universal behavior within the NFL regime at large $T/H^{3/2}$, for which we observe $\Gamma_H H \propto (T/H^{3/2})^{-4/3}$.

By expanding the dimensionless argument $(bh)/T^{1/\nu z}$ of the scaling function ϕ for high temperatures $(bh)/T^{1/\nu z} \ll 1$, we obtain $\Gamma_H H \sim (T/H^{\nu z})^{-2/\nu z}$ [27]. The experimentally observed high-temperature exponent $-4/3 = -2/\nu z$ (cf. dashed black line in Fig. 2) is perfectly consistent with $\nu z = 3/2$, obtained from the argument of the scaling function $\phi(T/H^{3/2})$. Previously, it has been shown that within the NFL regime, the "thermal" Grüneisen parameter given by the ratio of thermal expansion to specific heat diverges like $T^{-2/3}$ for $CeCoIn_5$ [9]. The quantum critical scaling predicts that this exponent equals $-1/(\nu z)$ [6,24]. Thus, $\nu z = 3/2$ is fully consistent with the thermal expansion study. By contrast, a previous specific heat study analyzed their data within the three-dimensional antiferromagnetic Hertz-Millis-Moriya (3D AF HMM) model for which $\nu z = 1$ [6]. However, about 70% of the total specific heat is nuclear in origin at 0.1 K and fields near 5 T [8]. Therefore, it seems very difficult to distinguish a $C/T \sim 1 - c\sqrt{T}$ dependence, predicted by the 3D AF HMM model [6] (c is a constant) from a saturation of C/T at low temperatures due to the crossover to Fermi-liquid behavior. The magnetic Grüneisen ratio is more sensitive to such a crossover, since at the critical field it would diverge as $1/T$ for the 3D AF HMM model.

A further constraint on the critical exponents characterizing quantum criticality in $CeCoIn_5$ is obtained from the magnetic Grüneisen behavior in the FL state following $\Gamma_H H \approx 0.85$. Since scaling predicts $\Gamma_H = -\nu(d - z)/(H - H_c)$ [6,24], i.e., $-\nu(d - z) \approx 0.85$, it follows that $\nu d \approx 0.65$, which provides a strong constraint for the nature of quantum criticality in this system.

In order to examine how close the quantum critical field (H_c) is to zero, we investigated the optimum collapse of the data in the scaling plot by tiny variations of H_c. The phenomenological function $f(x) = c_1/(x_1 + x^{4/3}) - c_2/(x_2 + x^2)$, shown in Fig. 2, fits very well the experimental data for the whole parameter range. Here, $x = T/(H - H_c)^{3/2}$, and c_1, c_2, x_1, and x_2 are fitting parameters ($H_c = 0$ in Fig. 2). The first term in f represents the expected behavior for $x \gg 1$ [27], while the second one is a phenomenological correction for the low x region. Inset (b) of Fig. 2 shows weighted mean-square deviation (σ^2) of the data from the phenomenological function as a function of H_c (see the definition of σ^2 in Ref. [27]). The minimum of σ^2 represents the best collapse and is located at $H_c = 0.06$ T. Importantly, the main contribution to σ^2 arises from the scattering of the data, while the quality of the collapse is subleading for small values of the critical field. In particular, the difference between the σ^2 values for 0.06 T and zero field is marginal, which justifies us to conclude upon zero-field quantum criticality in the system.

Our observation of zero-field quantum critical scaling sheds new light on the recent proposals of a field-induced QCP near 4 T (for $H \parallel [001]$), which has been obtained from linear extrapolation of the NFL-FL crossover in thermal expansion, magnetoresistance, and Hall effect measurements [11,19]. Clearly, the magnetic Grüneisen ratio, which is the most sensitive thermodynamic probe for a field-induced QCP, does not diverge in the approach of 4 T but in the approach of zero field (cf. insets of Fig. 2). If we thus exclude a QCP at 4 T, we need to demonstrate that all the previous measurements would also be compatible with zero-field quantum criticality. For this purpose, we consider the T-H phase diagram shown in Fig. 3, where the color coding represents the size of Γ_H/H in the normal state, which indicates the entropy accumulation due to quantum criticality [6,24]. Previous NFL to FL crossovers are included as circles, whose linear extrapolation would yield $H_c \approx 4$ T [11,19]. However, our $T/H^{3/2}$ scaling (Fig. 2) proves that such a linear extrapolation is not justified, since the temperature scale in the critical scaling regime does not depend linearly on H but rather superlinearly on $H^{3/2}$. The superlinear crossover between the NFL and FL states is indicated by the two dashed lines in the phase diagram, which correspond to the positions of the maxima and inflection points of the magnetic Grüneisen ratio data of Fig. 2. We note that the previously determined crossovers all lie between these two lines, indicating that

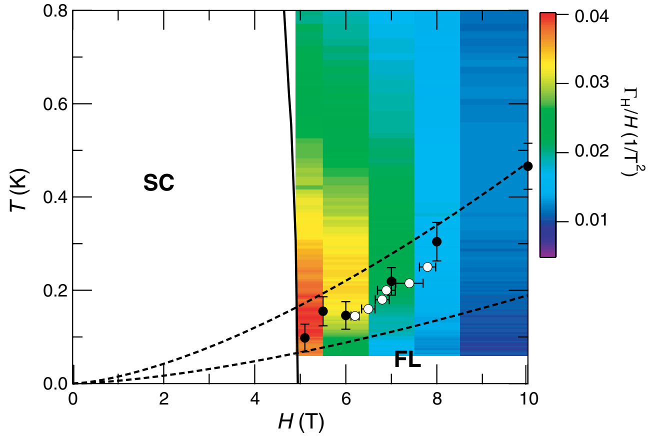

FIG. 3 (color online). T-H phase diagram of $CeCoIn_5$ for magnetic fields applied along [001]. The color code in the normal conducting state represents Γ_H/H. The onset of FL behavior determined by thermal expansion [11] (solid circles) and Hall effect [19] (open circles) measurements is included. The two broken lines represent the crossover between the NFL and FL regions $T_{\rm FL} = 0.015\ \mathrm{K\,T^{-3/2}}H^{3/2}$ and $T_{\rm FL} = 0.006\ \mathrm{K\,T^{-3/2}}H^{3/2}$, as defined by the maximum position and inflection point in $\Gamma_H H$, respectively, (see arrows in Fig. 2).

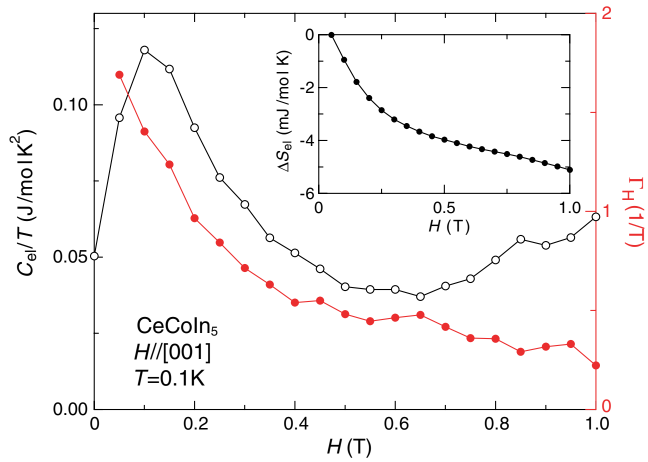

FIG. 4 (color online). Electronic specific heat divided by temperature $C_{\rm el}/T$ (open black circles, left axis) and magnetic Grüneisen ratio Γ_H (solid red circles, right axis) of $CeCoIn_5$ as a function of applied field along [001] at 0.1 K. The inset shows increments of isothermal electronic entropy as a function of field at 0.1 K.

these experiments are also compatible with zero-field quantum criticality.

In order to investigate signatures of zero-field quantum criticality on SC quasiparticles, we studied the low-field SC state by combined measurements of Γ_H and specific heat. Since $\partial S/\partial H = -C_{\rm el}\Gamma_H$, we obtain the isothermal field evolution of the entropy by integration. In our previous work, we focused attention near the SC upper critical field and found for $H \parallel [001]$ a broad kink in the field dependencies of Γ_H and $C_{\rm el}$ near 4.4 T [28]. Since this anomaly vanishes upon rotating the field towards the [100] direction, it cannot be related to the isotropic quantum critical scaling. We now concentrate on the behavior close to zero field. Figure 4 displays the measured heat capacity and magnetic Grüneisen ratio at low temperatures, together with the evolution of the entropy (see the inset). In nodal superconductors, quasiparticles exist at the gap nodes. Within the Shubnikov phase, the applied magnetic field creates vortices whose cores host additional quasiparticles. We therefore expect an increase of the entropy with increasing field for superconductors and a related negative sign of Γ_H. As shown in Fig. 4, remarkably, the magnetic Grüneisen ratio is positive and the entropy *decreases* with increasing field, contrary to the expectation for nodal superconductors and multiband superconductivity [29].

Relatedly, the isothermal field dependence of the specific heat coefficient $C_{\rm el}/T$ at 0.1 K (cf. Fig. 4) differs from the expected monotonic increase proportional to $\sqrt{H}$ for superconductors with line nodes of the gap [30,31]. The pronounced reduction of $C_{\rm el}/T$ with increasing field indicates that the quasiparticle mass is strongly enhanced near zero field. Thus, the peak in the field dependence of $C_{\rm el}/T$ arises from superposition of the usual $\sqrt{H}$ dependence with a quantum critical contribution. Near zero field, the latter vanishes due to the disappearance of vortices. Γ_H also shows a steep increase towards low fields consistent with zero-field quantum criticality. The anomalous decrease of quasiparticle entropy with increasing field (cf. inset of Fig. 4) is thus a consequence of this QCP near $H = 0$.

This is compatible with measurements of magnetic penetration depth, which found evidence for nodal quantum criticality at weak magnetic fields [32]. Previously, a pressure study of the SC upper critical field in $CeCoIn_5$ found a maximum in the SC coupling parameter near a pressure of 0.4 GPa, which has been interpreted as signature of a pressure-tuned QCP [33]. However, $H_{c2}(T)$ at low temperatures is also influenced by an unidentified high-field anomaly [28,34]. Therefore, such indirect conclusion on a QCP is rather uncertain.

To conclude, a systematic study of the magnetic Grüneisen ratio of $CeCoIn_5$ has revealed quantum critical scaling behavior in the normal state that indicates a critical field very close to zero in contrast to previous claims of a critical field only slightly below the upper critical field of superconductivity. The anomalous field dependence of the quasiparticle entropy within the SC state near zero field further supports this conclusion. It has been discussed in several classes of unconventional superconductors that T_c reaches its maximum near magnetic QCPs [1,3,35–39]. Since $CeCoIn_5$ exhibits nearly the highest $T_c = 2.3$ K among not only the 115 family but also all the structural variants of $Ce_nM_mIn_{3n+2m}$ (M, transition metal) [40], it is not surprising that $CeCoIn_5$ is located at a QCP without tuning any control parameters like composition or pressure or magnetic field. The observed scaling provides strong constraints on the critical exponents: $\nu z = 3/2$, consistent with previous thermal Grüneisen parameter studies [9], and $\nu d = 0.65$. Independent information of either d or z could then fully characterize quantum criticality in this system.

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