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 \( \Gamma_r = T^{-1}(dT/dr)_{\text{S}} \) (\( r \), pressure or magnetic field; \( S \), entropy), which diverge in the approach of the QCP [6]. Since magnetic field can easily be varied \textit{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] should be best 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.

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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 temperatures. Such temperature dependence is characteristic for fields above 6 T, saturates at lowest temperatures.

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, $\nu$ 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.

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

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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 $F_{c}$ for magnetic field as a tuning parameter can then be expressed by

$$F_{c} = aT^{(d+2)/\nu} \phi\left(\frac{bh}{T^{1/\nu}}\right),$$

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 $T/h^{\nu}$. All our $\Gamma_{h}$ 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 $\Gamma_{h}H = 0.85$ (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 $\phi$ for high temperatures $(bh)/T^{1/\nu_{z}} \ll 1$, we obtain $\Gamma_{h}H = (T/H^{\nu_{z}})^{-2/\nu_{z}}$ [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 $\Gamma_{h}H = 0.85$. Since scaling predicts $\Gamma_{h} = -\nu(d - z)/\nu(d - z) = 0.85$, it follows that $\nu d = 0.65$, 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 $f(x) = c_{1}/(x_{1} + x^{1/3}) - c_{2}/(x_{2} + x^{2})$, 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 ($\sigma^{2}$) of the data from the phenomenological function as a function of $H_{c}$ (see the definition of $\sigma^{2}$ in Ref. [27]). The minimum of $\sigma^{2}$ represents the best collapse and is located at $H_{c} = 0.06$ T. Importantly, the main contribution to $\sigma^{2}$ 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 $\sigma^{2}$ 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 $\Gamma_{h}/H$ 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 $H_{c} = 4$ 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
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 $\Gamma_H$ and specific heat. Since $\partial S/\partial H = -C_\text{el}\Gamma_H$, we obtain the isothermal field evolution of the entropy by integration. In our previous work, we focused attention near the SC state by combined measurements of $\Gamma_H$ and $C_\text{el}$ 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 $\Gamma_H$. 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 $C_\text{el}/T$ 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 $C_\text{el}/T$ with increasing field indicates that the quasiparticle mass is strongly enhanced near zero field. Thus, the peak in the field dependence of $C_\text{el}/T$ 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. $\Gamma_H$ 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$_xM_mIn_{1n+2m}$ ($M$, 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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