# Probing the Quantum Critical Behavior of $CeCoIn_5$ via Hall Effect Measurements

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We present highly sensitive Hall effect measurements of the heavy fermion compound $CeCoIn_5$ down to temperatures of 55 mK. A pronounced dip in the differential Hall coefficient $|\partial \rho_{xy}/\partial H|$ at low temperature and above the upper critical field of superconductivity, $H_{c2}$, is attributed to critical spin fluctuations associated with the departure from Landau Fermi liquid behavior. This identification is strongly supported by a systematic suppression of this feature at elevated pressures. The resulting crossover line in the field-temperature phase diagram favors a field induced quantum critical point at $\mu_0 H_{qc} \approx 4.1$ T below $H_{c2}(T = 0)$ suggesting related, yet separate, critical fields.

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Quantum criticality is a central issue of research in the physics of highly correlated electron materials. It is generally held that superconductivity (SC) in rare earth based heavy fermion (HF) metals occurs in the vicinity of a quantum critical point (QCP)—the point of transition between two stable phases at absolute zero temperature—and that this singularity organizes quite broadly the physics of this interesting class of materials. Naturally, this physics is expected to be relevant to a wide range of magnetic and almost magnetic materials, a span that includes high temperature superconductors. Detailed studies on high-quality HF materials at low temperature provide a particularly clean way to address key issues in the physics of QCPs. An important question concerns the evolution of the Fermi surface (FS) of a magnetically ordered metallic ground state at the QCP where the magnetic order is destroyed by quantum fluctuations. The Hall effect has been a particularly useful probe to address this issue. In $YbRh_2Si_2$, a recent Hall effect study [1] provided the first evidence for a discontinuous change of the FS across a QCP, consistent with the predicted disintegration of composite quasiparticles of the antiferromagnetic (AFM) HF ground state. It is highly desirable to extend such investigations to other strongly correlated metals that have a QCP in their phase diagram.

A ubiquitous issue in strongly correlated electron systems is to understand the relation between unconventional SC and quantum criticality. In many rare earth based compounds SC is found in the vicinity of an AFM QCP [2]. The QCP is located where the AFM ordering temperature, $T_N$, is suppressed to zero, suggesting that the superconducting state results from a magnetically mediated pairing mechanism. In this context, the tetragonal HF metals $CeMIn_5$ ($M$ = Co, Rh, Ir) provide an interesting playground. While $CeRhIn_5$ is one example of SC induced near an AFM QCP, $CeCoIn_5$ has a puzzling phase diagram which has been the focus of much attention since its discovery. It exhibits a magnetic field-tuned QCP [3,4] with a quantum critical field, $H_{qc}$, that appears to coincide with the *suppression* of SC at the upper critical field $H_{c2}^0 = H_{c2}(T = 0)$ at ambient pressure [5,6]. The origin of the QCP is still a matter of debate; a recent claim is that SC may conceal an AFM order [6].

In this Letter we present highly sensitive Hall effect measurements in $CeCoIn_5$ down to a temperature ($T$) of 55 mK. The quantum spin fluctuations (SF) appear to significantly influence the material's Hall response in a narrow low-$T$ range at ambient pressure, an effect which is gradually suppressed by applied pressure. In addition, these measurements indicate a dissociation of $H_{c2}^0$ and $H_{qc}$ suggesting that the SF may not become critical exactly at $H_{c2}^0$ but at a somewhat smaller field.

Hall voltages were measured by conducting isothermal field sweeps on three high-quality single crystalline $CeCoIn_5$ samples (resistivity $\rho \approx 1.4\ \mu\Omega$ cm at 2.4 K) with the crystallographic $c$ axis parallel to the applied field $H$, and with a sensitivity better than 0.05 nV (current $\leq$ 50 $\mu$A) [7]. For measurements under pressure $p \lesssim$ 1.2 GPa, the samples were mounted in a piston cylinder-type pressure cell using fluorinert 75 as the pressure transmitting medium. Results of the Hall measurements at various temperatures and $p = 0$ are shown in Fig. 1 (for clarity, not all isotherms are presented). Our Hall resistivity, $\rho_{xy}(H)$, is negative corresponding to an electron dominated transport, and can favorably be compared to previous reports above 1 K [8–10]. Here, however, we concentrate on the low-$T$ regime, $T \leq 0.32$ K. In this regime, the overall behavior of $\rho_{xy}(H)$ changes appreciably with $T$. In addition, $\rho_{xy}(H)$ is nonlinear in $H$; *only* at the lowest $T$ is linearity recovered, with $\rho_{xy}(H)$ extrapolating to zero at $H = 0$ [Fig. 1(b), dashed line]. The magnetization $M(H)$ in the normal state shows an almost linear dependence on $H$ and very little $T$ dependence for 67 mK $\leq T \leq$ 0.37 K [11,12], and hence the peculiarities in $\rho_{xy}(H)$ cannot be attributed to changes in $M(H)$. In addition, our $T$ range is well below the coherence temperature $T^* \sim 40$ K [9,13]. Furthermore, the anomalous contribution due to skew scat-

 

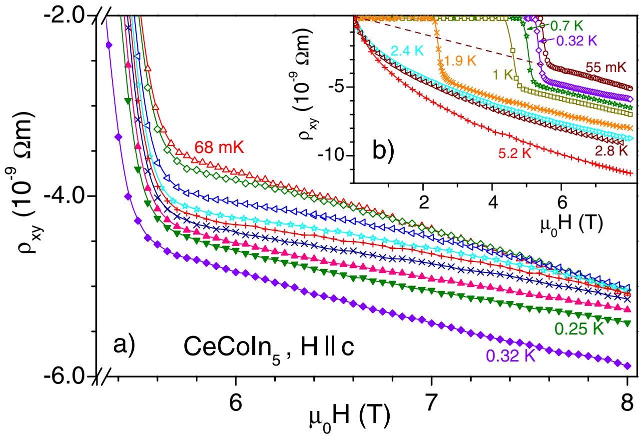


FIG. 1 (color online). Isothermal Hall resistivity $\rho_{xy}$ as a function of $H$ for $CeCoIn_5$ for selected temperatures. (a) Low temperature range $T \leq 0.32$ K showing the onset of SC (curves not labeled: $T = 0.09$, 0.115, 0.145, 0.16, 0.18, 0.215 K). For $T \leq 0.25$ K, $T$-dependent changes in slopes of $\rho_{xy}(H)$ are observed in the normal-conducting regime. (b) $T$ range up to 5.2 K. The dashed line extrapolates $\rho_{xy}(H, T = 55 \text{ mK})$ to $H = 0$.

tering was shown to be small for $T < 50$ K [8,9]. Therefore, we can safely assume that $R_H = \rho_{xy}/\mu_0 H$ mainly probes the FS volume of $CeCoIn_5$ at temperatures of interest here.

Figure 2(a) shows $|R_H|$ at $T \leq 0.32$ K and ambient pressure as a function of $H$. Interestingly, the $|R_H|$ data form part of a single generic curve when their field dependences are scaled by $H_{\min} \propto 1/\mu_{\text{eff}}$; see Fig. 2(b) (only data not influenced by the transition into the superconducting phase, i.e., well above $H_{c2}$, are shown). Here, $H_{\min}$ corresponds to the field at which $|R_H|$ assumes a minimum and $\mu_{\text{eff}}$ can be interpreted as an effective carrier mobility averaged over the different sheets of the FS contributing to the Hall voltage. For 70 mK $\leq T \leq$ 180 mK a minimum in $|R_H|$ is directly observed. With increasing $T$ this minimum shifts to higher $H$ and is no longer observed for $T \geq$ 0.2 K within our accessible field range; i.e., the value of $H_{\min}$ can only be estimated from the scaling. This scaling suggests that $\mu_{\text{eff}}$ is primarily a function of $T$, with only a weak field dependence. Obviously, $\rho_{xy}(H)$ does not obey a linear $H$ dependence; therefore, the differential Hall coefficient, $R_H^d = \partial \rho_{xy}(H, T)/\partial H$, is a quantity of interest and the same scaling used for $R_H$ works for $R_H^d$ with identical $H_{\min}(T)$ [Fig. 3(e)].

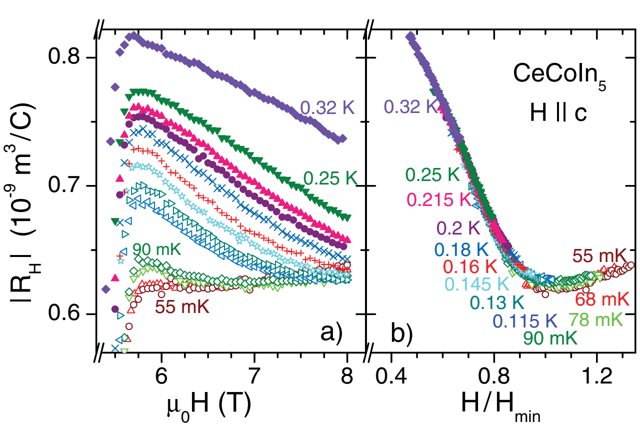


FIG. 2 (color). (a) Hall coefficient $|R_H| = |\rho_{xy}|/H$ of $CeCoIn_5$ at $T \leq 0.32$ K. (b) Same $|R_H|$, but $H$ values are scaled by $H_{\min}$ to achieve maximum overlap of the curves. For clarity, data influenced by transition into SC [as seen in (a)] are not shown.

The scaling of $|R_H^d|$ at $p = 0$ reveals a prominent minimum (dip) corresponding to the dent in $|R_H|$. We note that the minimum value of $|R_H^d|$ at a scaled field $H_d(T) \approx 0.8H/H_{\min}$ is only *half* of that outside the dip and would correspond to a doubling of the carrier density $n$ if $n \propto 1/R_H^d$ is assumed. The dip appears on top of a baseline which exhibits only a weak field dependence within the investigated field range: $R_H^d \approx -0.6$ m$^3$/C for $H < 0.55H_{\min}$ and $\approx -0.7$ m$^3$/C for $H > 1.1H_{\min}$. Moreover, at 60 mK the same value persists up to a pressure of 1.2 GPa [Fig. 3(a)]. It is important to note that these values agree well with those reported for the nonmagnetic analog $LaCoIn_5$ [10]. From Hall data above 1.8 K, $R_H^d$ of $CeCoIn_5$ in zero-field limit was found [10] to approach that of $LaCoIn_5$ with increasing pressure. Since resistivity at $H = 0$ indicates a crossover to a Landau Fermi liquid (LFL) regime at high pressure [14], it was speculated that the $R_H^d$ value of $LaCoIn_5$ corresponds to the LFL value for $CeCoIn_5$. Following the same line of argument, one may interpret the baseline value as an indication of LFL behavior—with the marked exception of the dip feature. The fact that $\rho_{xy}(H)$ at 55 mK extrapolates to zero for $H = 0$

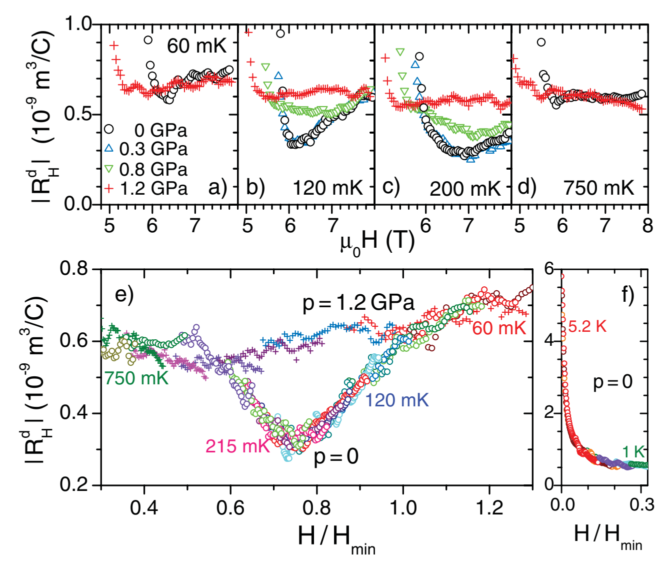


FIG. 3 (color). Comparison of $R_H^d$ obtained at different pressures and for (a) $T = 0.06$, (b) 0.12, (c) 0.2, and (d) 0.75 K. The evolution of the low-$T$ Hall feature at low pressure is obvious. (e) The scaled $|R_H^d|$ at $p = 0$ (○) show a pronounced dip feature which is nearly suppressed at $p = 1.2$ GPa (+). Again, data influenced by the transition into SC are omitted. (f) Scaled $|R_H^d|$ at $p = 0$ for $T \geq 1.0$ K.

[dashed line in Fig. 1(b)] further supports this assumption as the corresponding $R_H^d$ data completely fall onto the baseline on the high-field side of $H_{\rm min}$.

For small fields $H \lesssim 0.2H_{\rm min}$ and for $T > 1$ K, a steep increase of $|R_H^d|$ with decreasing $H$ is observed [Fig. 3(f)] so that our data agree with those reported for $T \gtrsim 2$ K [8]. A rough estimate from the resistivity yields $\omega_c\tau \gtrsim 1$ at 5 T and 0.3 K ($\omega_c$, cyclotron frequency; $\tau^{-1}$, scattering rate). Hence, we interpret the increase of $|R_H^d|$ for $H \leq 0.2H_{\rm min}$ as a departure from the high-field regime.

Nuclear quadrupole resonance measurements in $\rm CeCoIn_5$ [15,16] have revealed AFM SF in the normal state with $1/T_1 \sim T^{1/4}$ indicating the proximity to a QCP. Additionally, transport and specific heat ($C_p$) data [3,4] point to a field-tuned QCP close to $H_{c2}^0$, with a possible AFM ground state superseded by SC. Hence, it is tempting to investigate the relationship between the SF and the dip feature in $R_H^d$. For increasing pressure a tendency toward a LFL state has been observed [10,14,17,18], concomitant with the suppression of SF [19]. Therefore, we measured the Hall Effect under pressure up to $p = 1.2$ GPa at low $T$. Figures 3(a)–3(d) present the field dependence of $|R_H^d|$ at different $p$. We observe a decreasing $H_{c2}$ with increasing pressure, in good agreement with previous reports [14,17,20]. For $T$ below (60 mK) and above (750 mK) the dip feature, pressure has little effect on $|R_H^d|$. In contrast, the dip feature itself is gradually abated with increasing $p$, as shown for 120 and 200 mK in Figs. 3(b) and 3(c), with no significant changes between $p = 0$ and 0.3 GPa. The overall behavior is evident in Fig. 3(e) where the field-scaled Hall data measured at $p = 0$ and 1.2 GPa are compared. Here, the $T$ dependence of $H_{\rm min}$ at ambient pressure was used for the scaling of the $H$ values of the Hall data at 1.2 GPa. The scaling works less well when pressure is applied. For $p = 0.8$ GPa (not shown), we cannot collapse the $R_H(T)$ [or $R_H^d(T)$] data to a single curve; i.e., $H_{\rm min}(T)$ does not capture the $T$ dependence of the dip feature in $R_H^d$, although the baseline stays intact. Even though the dip feature appears to be strongly reduced at 1.2 GPa, it may not be completely suppressed. Note that this pressure is very close to the one where $H_{qc} \rightarrow 0$ [18]. Our results, along with the magnetoresistance (MR) [18], suggest that the minimum feature in $|R_H^d|$ is related to AFM SF. Moreover, the above-mentioned baseline on which this dip evolves does not show significant pressure dependence. de Haas–van Alphen (dHvA) measurements observed a nearly unchanged FS topology with increasing pressure [17], which we consider as a further indication that the baseline in $R_H^d$ represents a LFL regime.

To further pursue the idea that the dip feature may be related to strong AFM SF, a $T$-$H$ phase diagram constructed from the Hall effect is presented in Fig. 4. The field range corresponding to the superconducting transition in $\rho_{xy}(H)$, and consequently $R_H^d$, is marked by the hatched area. The onset of the transition, $H_{c2}^{\rm Hall}(T)$, agrees well with values of $H_{c2}(T)$ obtained from other measurements [21]. A broad

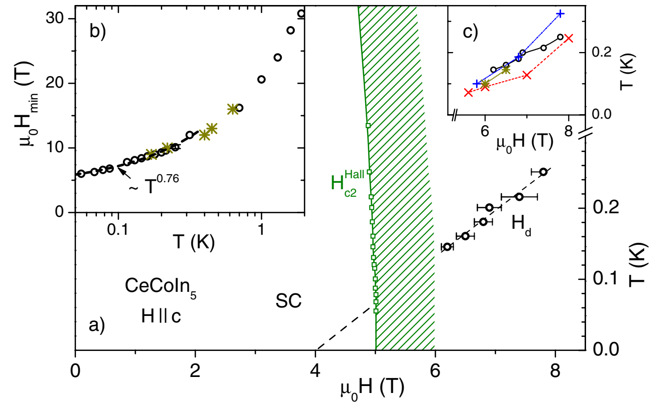


FIG. 4 (color online). (a) Phase diagram resulting from Hall effect measurements. The hatched area marks the $H$ range within which SC influences the slope of $\rho_{xy}(H)$ [□ mark onset of nonzero $\rho_{xy}(H)$]. $H_d(T)$ values for which a minimum in $|R_H^d|$ can directly be observed are marked by ○. A power law fit extrapolates to 4.1 T at $T = 0$ (dashed line). (b) $T$ dependence of $H_{\rm min}$. A fit (dashed line) again extrapolates to 4.0 T at $T = 0$. MR data (*) taken from Ref. [3]. (c) Our results compared to those from resistivity and specific heat measurements in Refs. [6] (+, ×) and [3] (*). Lines are guides to the eye.

transition into SC is commonly observed in $\rho(H)$ [22] and may be due to superconducting fluctuations. For temperatures where the minimum in $|R_H^d|$ can be directly observed $H_d(T)$ is marked (○). In Fig. 4(c) these are compared to the crossover to LFL behavior in $\rho(T)$ and $C_p(T)$ from Refs. [3,6] and indicate very good agreement. For $T \leq$ 130 mK the dip feature may still exist but be masked by the approach to SC. When $H_d(T)$ is extrapolated to $T \rightarrow 0$ by a simple power law, we find $\mu_0 H_d(0) = 4.1(+0.8; -2.2)$ T. In Fig. 4(b), the $T$ dependence of $H_{\rm min}$ resulting from the scaling in Fig. 2 is presented. $H_{\rm min}$ compares favorably to the crossover from negative to positive MR [3,8]. The $H_{\rm min}$ data at $T \leq 0.32$ K can also be fit well by a power law $H_{\rm min} = a + bT^c$ with $a = 4.0 \pm 0.7$ T and $c = 0.76 \pm 0.13$. The following facts further support our scaling: (i) fitting $H_{\rm min}$ data up to $T = 5.2$ K yields very similar parameters, (ii) $a$ is very close to $H_d(0)$, and (iii) $c$ agrees well with the exponent of the divergence in $A(H)$ [3], the $T^2$ coefficient of $\rho = \rho_0 + AT^2$, and the scaling exponent of $C_p$ [4]. While the determination of $A(H)$ suffers from the presence of an upturn in $\rho$ at the lowest $T$ [3,6,18], our method based on the $H$ dependence of $R_H^d$ avoids this problem and may apply to the MR data as well.

There is no stringent criteria for the LFL regime in the Hall effect as, e.g., the $T^2$ law in resistivity. Nonetheless, the comparison to $\rho_{xx}$ and $C_p(T)$ data [Fig. 4(c)] suggests that the dip feature in $R_H^d$ may be related to the crossover from LFL to non-Fermi liquid (NFL) regime [4]. Thus, $H_d(T)$ is bound to approach the quantum critical field $H_{qc}$ as $T \rightarrow 0$. Our measurements of $C_p(T, H)$ point to $H_{c2}^0 =$ $4.95 \pm 0.07$ T, in concert with Ref. [21]. Hence, $\mu_0 H_{qc} =$ 4.1 T is likely smaller than $H_{c2}^0$, as suggested by recent MR measurements [18,22], but in contrast to the conclusion drawn in Refs. [5,6] that $H_{qc} = H_{c2}^0$. Even though our error of $H_{qc}$ does not completely rule out the case $H_{qc} = H_{c2}^0$, this would require an unusually large exponent of 1.3 in the power law fit of $H_d$ [Fig. 4(a)] and is outside the error of $H_{min}(0)$. Our $H_{qc}$ seems also distinct from the onset of the Fulde-Ferrell-Larkin-Ovchinnikov state at $H > 4.7$ T recently claimed for $H \parallel c$ [23]. In view of a possible existence of a group of electrons that is not involved in SC but may participate in magnetism and quantum criticality [24], a certain autonomy of $H_{c2}^0$ and $H_{qc}$ is naturally supported. Moreover, our scaling form $R_H = f(\frac{H}{a+bT^c})$ is similar to the one reported [9] for $T > T_c$, the critical temperature of SC. The emergence of the same energy scale [$T_K \approx 1.7$ K or $H_{min}(0) = 4.0$ T] in these low- and high-field scalings may suggest an unprecedented connection between the origin of the field-tuned QCP and the single-ion Kondo scale $T_K$. If so, one might speculate that $H_{qc}$ being close but not identical to $H_{c2}^0$ is due to the proximity of $T_K$ and $T_c$ in this material.

Hall effect measurements can be used to address the nature of a QCP (local or spin density wave type) [1], via the inference of a jump in the Hall constant at $T = 0$ in the case that the FS volume changes abruptly. In the present case SC prevents a comparison of the FS volume across the QCP. The dip feature in $R_H^d$ is reminiscent of the anomaly recently predicted for a FS topology change in the spin density wave scenario [25] which is likely applicable to $CeCoIn_5$ [26]. However, the pressure suppression of the dip disfavors such an explanation, since the FS changes only slightly under pressure [17]. On the other hand, the similar $H$ dependence observed in $|R_H|$ and the MR [3,8] within our field range (both increase for the lowest $T$ but decrease for $T \gtrsim 300$ mK) is consistent with a two-band model where the carrier density increases with $H$ at constant mobility $\mu$. This suggests that the $f$ electrons become more itinerant with increasing $H$, in agreement with a putative QCP. A nearly $H$-independent $\mu$ is consistent with our scaling in Fig. 2. Note that the renormalization of the effective mass $m^*$ and of the scattering time $\tau^*$ cancel each other in $\mu = \frac{e\tau^*}{m^*}$ at the mean field level [27]. It is also possible that the individual mobilities for electrons and holes depend on $H$ [17], but nearly compensate each other in FS-averaged $\mu$. In fact, band structure calculations [28,29] as well as dHvA data [29,30] reveal $CeCoIn_5$ to be a nearly compensated metal with multiple bands at the Fermi level. This may result in the slightly $H$-dependent baseline displayed by our data.

In conclusion, the Hall coefficient of $CeCoIn_5$ has a distinct minimum between 115 mK $\leq T \leq$ 250 mK which can be suppressed by applying pressure. The pressure evolution of $R_H^d$, combined with the value of $R_H^d$ corresponding to the expected LFL value at the lowest $T$ and the comparison of the characteristic field $H_d(T)$ to the known phase diagram at $p = 0$, suggests that this feature is related to the crossover from NFL to LFL behavior. Thus, our data support the idea of a field-tuned QCP in $CeCoIn_5$ which is not exactly located at $H_{c2}^0$. Rather, it is within the field range of SC which appears to mask an AFM ordered state generating this QCP. We believe that this disparity does not necessarily rule out the possibility of the AFM SF being involved in the *formation* of $d$-wave SC, even if they do not become critical right at $H_{c2}^0$. The similarity between $R_H$ and MR indicate that a similar scaling might prevail in the MR. Finally, the dissociation between $H_{qc}$ and $H_{c2}^0$ as well as the likely emergence of the single-ion Kondo scale in the high-field region of the phase diagram may be an important observation for untangling the origin of the QCP in $CeCoIn_5$.

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