Avoided Antiferromagnetic Order and Quantum Critical Point in CeCoIn$_5$

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We measured the specific heat and resistivity of heavy fermion CeCoIn$_5$ between the superconducting critical field $H_{c2} = 5$ T and 9 T, with the field in the [001] direction, and at temperatures down to 50 mK. At 5 T the data show a non-Fermi liquid (NFL) behavior down to the lowest temperatures. At the field above 8 T the data exhibit a crossover from the Fermi liquid to a non-Fermi liquid behavior. We analyzed the scaling properties of the specific heat and compared both the resistivity and the specific heat with the predictions of a spin-fluctuation theory. Our analysis leads us to suggest that the NFL behavior is due to incipient antiferromagnetism (AFM) in CeCoIn$_5$ with the quantum critical point in the vicinity of H_{c2}. Below H_{c2} the AFM phase which competes with the paramagnetic ground state is superseded by the superconducting transition.

When the symmetry of the ground state of a system changes as a function of an external or internal parameter, the system is said to undergo a quantum phase transition. If, in addition, this transition is second order, the system has a quantum critical point (QCP) at the critical value of the parameter. The competition between the nearly degenerate ground states determines the behavior of the system over a range of temperatures and tuning parameter values in the vicinity of QCP. In this region of the phase diagram the properties of the system differ from those on either side of the transition and often exhibit unusual dependence on the temperature and the tuning parameter. This has made quantum critical phenomena a subject of intense current interest.

The study of quantum critical points in heavy fermion systems has been the focus of particular attention (for a recent review see Ref. [1]). In these materials the competition typically takes place between a paramagnetic and a magnetically ordered ground state. The unconventional behavior near a QCP is manifested in the deviation of the temperature dependence of measured properties from those of metals described by the Landau Fermi liquid (FL) theory. In that theory the electronic specific heat is linear in temperature, $C(T) = \gamma T$, and the resistivity increases quadratically from a residual value, $\rho = \rho_0 + AT^2$. In systems tuned to a QCP, the Sommerfeld coefficient $\gamma(T) = C/T$ commonly diverges as the temperature goes to zero and has been variously argued to behave as either $\log T$ or T^α, with $\alpha < 0$. A resistivity with an exponent less than 2 is also ubiquitous in these compounds.

Tuning the system through a QCP can be accomplished experimentally by varying the sample's composition [2,3], applying pressure [4], or applying a magnetic field [5]. In nonstoichiometric compounds the Kondo disorder, where a range of Kondo temperatures T_K appears due to different environments of the f-electron ions, is an important mechanism leading to a non-Fermi liquid (NFL) behavior [6,7]. In these compounds it is not easy to sepa-

rate this origin of NFL behavior from the consequences of the proximity to a QCP. Hence the stoichiometric compounds receive a great deal of attention in the field of quantum criticality.

One class of such materials is Ce-based compounds, which have an antiferromagnetic (AFM) ground state at ambient pressure. The hydrostatic pressure suppresses the magnetic ordering temperature T_N to zero at a critical pressure of the QCP. Such an approach was used successfully for CeCu$_2$Ge$_2$ [8], CeRh$_2$Si$_2$ [9], CePd$_2$Si$_2$ and CeIn$_3$ [4], and some other compounds. Alternatively, AFM order can be suppressed by an applied magnetic field. When this was done in YbRh$_2$Si$_2$, an NFL behavior was revealed again on the paramagnetic side of the QCP [10].

In this Letter we present the results of the specific heat and resistivity measurements on stoichiometric CeCoIn$_5$, which show that this compound has a QCP with a magnetic field as a tuning parameter. We show that the quantum critical behavior is most likely due to the proximity of an antiferromagnetic state. This case is particularly interesting because CeCoIn$_5$ is an ambient pressure superconductor. We argue that magnetic order that competes with the paramagnetic state is superseded by the onset of superconductivity.

CeCoIn$_5$ is a tetragonal, quasi-2D compound, with layers of CeIn$_3$ separated by layers of CoIn$_2$. It is an ambient pressure heavy fermion superconductor [11] with $T_c = 2.3$ K, the highest value for this class of compounds. Superconductivity in CeCoIn$_5$ is unconventional, with lines of nodes in the energy gap, as demonstrated by specific heat and thermal conductivity [12] and nuclear quadrupole resonance [13] measurements. A Pauli limiting analysis [14] and thermal conductivity modulations in magnetic field [15] indicate that CeCoIn$_5$ is a singlet, $d_{x^2-y^2}$ superconductor. Recently it was shown that the superconducting transition in CeCoIn$_5$ in magnetic fields close to the upper critical field for the [001] direction, $H_{c2} = 4.95$ T, is first order below $T = 0.7$ K [15,16]. NFL

behavior of CeCoIn$_5$ at the field of 5 T persists over a large region of temperature [17], with $\gamma \propto -\log(T)$ between 0.4 and 8 K.

Here we concentrate on the low temperature region down to 50 mK and magnetic fields between $H_{c2} = 4.95$ T and 9 T. Figure 1(a) shows a Sommerfeld coefficient of CeCoIn$_5$ versus temperature for several magnetic fields between 5 and 9 T. Large low temperature Schottky anomaly tails due to the quadrupolar and magnetic spin splitting of In and Co nuclei, contributing 70% and 95% of the total specific heat at 100 mK for 5 and 9 T, respectively, were subtracted [12]. At high magnetic field and low temperature the Schottky term accounts for most of the specific heat measured, and its subtraction results in an increased scatter of the data points at low temperature. The data for 5 T follow logarithmic temperature behavior below 1 K down to the lowest temperature
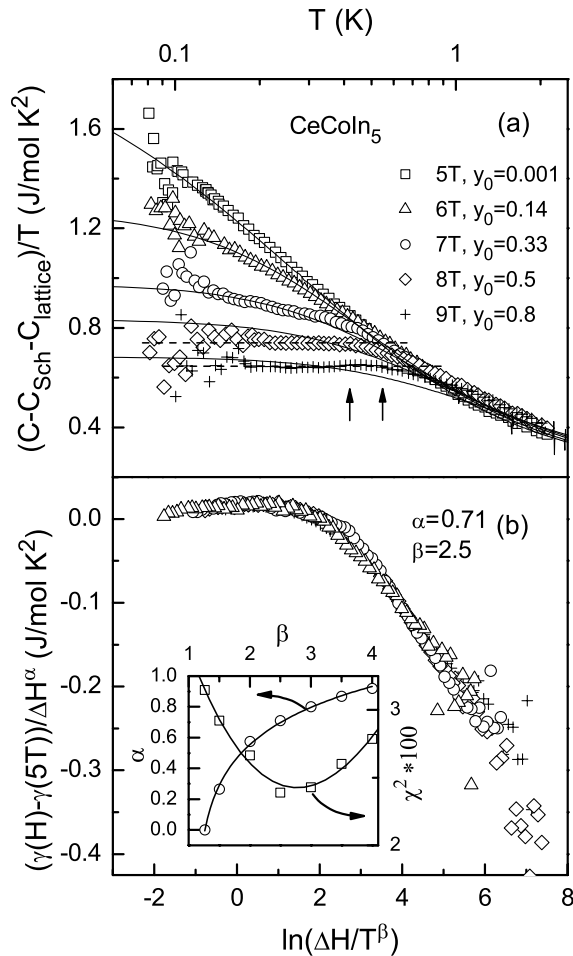

FIG. 1. (a) Sommerfeld coefficient, $\gamma(T) = C(T)/T$, of CeCoIn$_5$ in magnetic fields $H \parallel [001]$. Dashed lines for 8 and 9 T emphasize the FL behavior with constant γ. Left (right) arrows indicate T_{FL}^C for 8 T (9 T). Solid lines are fits to the SCR spin-fluctuation model for each field with the corresponding values of y_0; see text for details. (b) Scaling analysis of the data in (a) for $\alpha = 0.71$ and $\beta = 2.5$. Inset: For different values of β we plot values of α which minimize χ^2 for a given β, and χ^2 for these α and β.

studied. As the magnetic field is increased above 5 T, the low temperature data start to deviate from the logarithmic behavior at an ever higher temperature. A clear FL regime of $\gamma = $ const at low temperature is recovered at 8 T. Both 8 and 9 T data exhibit sharp crossovers between the NFL (logarithmic) and the FL regimes at a temperature T_{FL}^C. For fields below 8 T γ continues to rise with decreasing temperature and does not show saturation within the temperature range shown. This behavior is suggestive of a field-tuned QCP near 5 T. Indeed, if we were to shift all the field labels by -5 T, Fig. 1(a) would describe specific heat of a prototypical compound at a zero-field quantum critical point, such as U$_{0.2}$Y$_{0.8}$Pd$_3$ [18] and YbRh$_2$Si$_2$ [19].

This observation is strongly supported by the scaling properties of γ. For a phase transition with $T_c = 0$, the temperature alone sets the energy scale of fluctuations. Scaling of dynamical properties with energy, E, as E/T is taken as good evidence of quantum criticality [20,21]. In our case the tuning parameter is the applied magnetic field. Hence the energy scale of fluctuations away from the QCP depends on $|H - H_c|$, and the scaling parameter is $(H - H_c)/T^\beta$, where β is the scaling dimension of the magnetic field. Following the argument of Ref. [22] we arrive at the scaling function of the form $\gamma(H) - \gamma(H_c) \propto (H - H_c)^\alpha f((H - H_c)/T^\beta)$. The form of scaling is similar to that obtained for U$_{0.2}$Y$_{0.8}$Pd$_3$ [18], CeCu$_{5.8}$Ag$_{0.8}$ [23], and for YbRh$_2$Si$_2$ [19].

We find that with the choice of $H_c = 5$ T, the best scaling is achieved for $\alpha = 0.71$ and $\beta = 2.5$; it is shown in Fig. 1(b). It is a remarkably good scaling, spanning both FL and NFL regimes, with all four data sets for different fields overlapping each other in the entire experimental temperature range. This scaling is a strong indication that the behavior of CeCoIn$_5$ in the part of the phase space explored in these experiments is governed by the QCP very close to $H_c = 5$ T. The high power of temperature in the argument of the scaling function (the scaling dimension of the magnetic field) suggests that the field is very efficient in suppressing the critical fluctuations.

Importantly, scaling implies that the behavior observed here is associated with a second order transition. Therefore it is unlikely that first order transition from the superconducting to normal state above 4.7 T [16] controls the properties in the parameter range investigated here, and we need to consider alternative competing orders. We argue below that critical AFM fluctuations lead to the observed NFL behavior in CeCoIn$_5$.

The crossover from the NFL to the FL regime is also clear from resistivity measurements in the same magnetic field range. Figure 2(a) shows resistivity, ρ, of CeCoIn$_5$ below 5 K for fields between 5.6 and 9 T, as well as the zero-field data. The data at 5.6 T and above are not affected by the proximity to the superconducting transition, as seen from the magnetoresistance measurements of CeCoIn$_5$ at 100 mK [24]. To focus on the low temperature behavior, we show the data below 1 K in
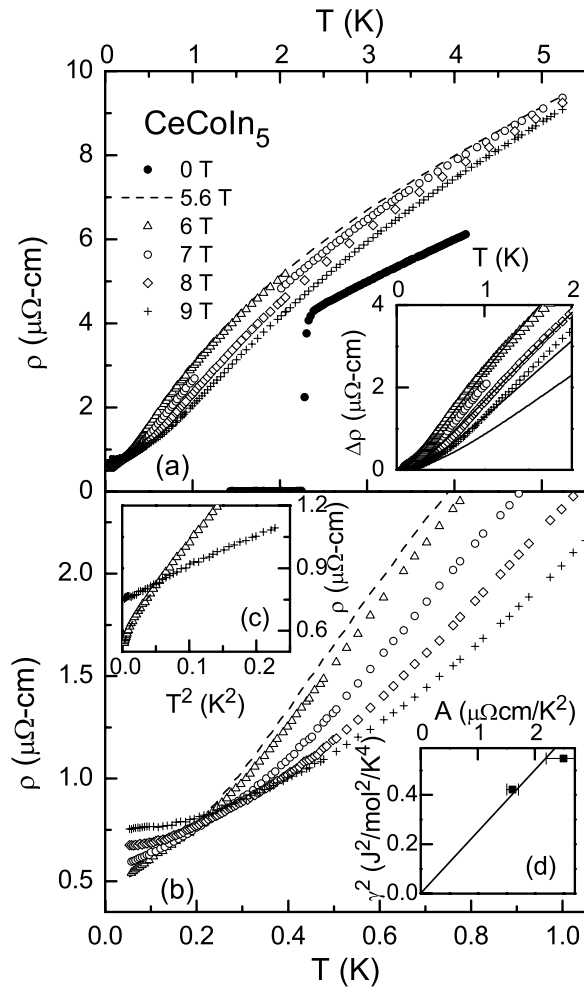
FIG. 2.   (a) Resistivity of CeCoIn$_5$ in magnetic field $H \parallel$ [001] above the critical field $H_{c2} = 4.95$ T. Inset: Fits to $\Delta\rho(T)$ below 2 K from the spin-fluctuation theory as detailed in the text. Solid lines: $y_0 = 0.14$, 0.33, 0.5, and 0.8 from top to bottom. (b) Low temperature region of data in (a). Inset (d) γ^2 vs A. The solid line is a linear fit to the data points and the origin. Inset (c) Resistance for 6 and 9 T plotted vs T^2. All symbols are as indicated in (a).

Fig. 2(b). The data for 8 T below 300 mK and for 9 T below 500 mK display the FL-like $\Delta\rho \equiv \rho - \rho(T = 0) = AT^2$ behavior, consistent with the linear in T specific heat data for these fields. In contrast, for fields of 5.6 and 6 T we found $\Delta\rho \propto T$ below 200 mK, and the data for 7 T show intermediate behavior, the best fit being the sum of linear and quadratic terms in this temperature range. At these fields the linear in T, rather than quadratic, dependence of the resistivity is consistent with the NFL behavior of the specific heat. A qualitative difference in the temperature behavior of $\rho(T)$ at 6 and at 9 T in the low temperature range is emphasized in the upper inset of Fig. 2(b), where resistivity is plotted versus T^2 for two fields, 9 and 6 T. The 9 T data are linear in T^2 over the entire temperature range shown, while the 6 T data have a pronounced negative curvature.

To further analyze the FL region of the phase diagram of CeCoIn$_5$, in the inset (d) of Fig. 2(b) we plot γ^2 versus

A for the 8 and 9 T data. Within error bars the two values lie on the line $\gamma^2 = 0.3 \times A$ J^2/mol^2 K^2 $\mu\Omega$ cm. The slope is about 3 times larger than the Kadowaki-Woods ratio [25], approximately obeyed by many heavy fermion compounds. This discrepancy is within the typical range of the scatter of the data for other HF compounds and confirms the FL ground state of CeCoIn$_5$ for 8 and 9 T.

Having established the existence of the QCP near $H_c \approx$ 5 T, we proceed to identify the order competing with the paramagnetic FL ground state. Since a closely related compound, CeRhIn$_5$, is an ambient pressure antiferromagnet with the Néel temperature $T_N = 3.8$ K, AFM order is a natural candidate. It is also consistent with the $d_{x^2-y^2}$ superconductivity.

While the complete theory of quantum criticality in itinerant antiferromagnets is still lacking, the singular contribution of the critical AFM spin fluctuations to the specific heat and scattering has been considered in Ref. [26] in the framework of a self-consistent renormalization (SCR) theory. This theory was used to analyze the specific heat of CeCu$_{5.2}$Ag$_{0.8}$ near the QCP [1]. The input parameter of the theory is the distance from a QCP, y_0, ($y_0 = 0$ at the QCP), related to the inverse square of the magnetic correlation length. The specific heat and the resistivity are functions of the reduced temperature, T/T_0, where $2\pi^2 T_0$ is of the order of the exchange interaction. We use this theory to fit our data.

Close to the QCP we obtained a good fit to both the specific heat data [shown in Fig. 1(a)], and resistivity [shown in the inset of Fig. 2(a)] for the same parameter values. To estimate T_0, we compared CeCoIn$_5$ with CeRhIn$_5$. Pressure of 16 kbars suppresses the AFM state in CeRhIn$_5$, at which point the superconducting state with $T_c = 2.1$ K emerges [27]. This led Sidorov et $al.$ to suggest that physical properties of CeCoIn$_5$ are very close to those of CeRhIn$_5$ at 16 kbars [28]. The $T_0 = 0.4$ K we chose is consistent with the exchange energy of the order of a few Kelvin, expected from the $T_N = 3.8$ K for CeRhIn$_5$. We fit the data to a sum of an SCR term [26] and a field-independent contribution from noncritical fermions, which turns out to be $\gamma(T) = 0.2$ J/mol K^2. For the 5 T data the theoretical fits are identical for any $y_0 \leq 0.01$ over the temperature range studied. Consequently, CeCoIn$_5$ is very close to a QCP at this field.

Since in our experiments the magnitude of the applied field (used to tune the system to the QCP) is comparable to the exchange interaction, we expect that the changes in the spectrum of the spin fluctuations and the quasiparticle scattering rate lead to deviations from the predictions of the SCR theory when the field is increased away from QCP. Indeed, at higher fields the discrepancy between the fits and the resistance data becomes more pronounced (especially at higher temperature). However, the overall S shape of the resistance curves agrees with the data. We therefore conjecture that the QCP is due to the tendency towards the AFM order.
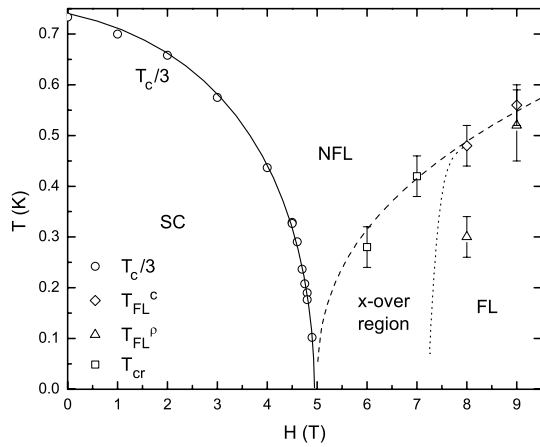
FIG. 3. Combined phase diagram of CeCoIn$_5$. Superconducting-normal phase boundary [($\bigcirc$) and solid line] is from Ref. [16]. T_{cr} ($\square$) and the dashed line (guide to the eye) denote the upper boundary of the crossover region. Dotted lines through T_{FL}^C ($\diamond$) and T_{FL}^ρ ($\triangle$) schematically indicate the upper boundary of the FL region. The region between the dashed and the dotted lines is the crossover region.

Finally, in Fig. 3 we plot the phase diagram of CeCoIn$_5$ in the T-H plane. The normal-superconducting phase boundary was determined in Refs. [11,16]. At fields of 8 and 9 T we see a clear transition from the NFL to the FL behavior in both specific heat (T_{FL}^C) and resistivity (T_{FL}^ρ). Below 8 T, in the temperature range studied, the heat capacity and resistance deviate from the critical behavior (5 T data), although the FL regime is not reached. For these fields we define a temperature T_{cr} below which γ begins to deviate from $-\log(T)$ behavior, leading to a broad crossover region. If the FL behavior exists at these fields, it occurs over a narrow range of very low temperatures.

In conclusion we find that the behavior of CeCoIn$_5$ above the superconducting upper critical field, $H_{c2} = 4.95$ T, at low temperatures is controlled by a field-tuned antiferromagnetic quantum critical point. The existence of the QCP is confirmed by a scaling analysis of the specific data. It is also indicated by the susceptibility [29] of CeCoIn$_5$, which follows a modified Curie-Weiss law observed in other systems near QCP's. An incipient AFM state in CeCoIn$_5$ is strongly suggested by the fits to both the specific heat and the resistivity based on the spin-fluctuation theory [26] with AFM fluctuations becoming critical at fields close to 5 T. The underlying physical picture is that the AFM spin fluctuations promote anti-alignment of the electron spins. Whether time-reversal breaking (antiferromagnetism) or singlet formation with phase coherence (superconductivity) occurs first depends on the details of the system. In CeCoIn$_5$ the superconductivity prevails, preventing the magnetic order from developing. However, there are no critical fluctuations associated with the (first order [16]) superconducting transition at H_{c2}, so that the behavior above H_{c2} is controlled by the proximity to the AFM critical point. Our results show that the antiferromagnetic fluctuations become critical in the immediate vicinity of the superconducting upper critical field. This concomitant suppression of the AFM and superconducting orders is unusual and may reflect an important aspect of the underlying physics, presenting further challenges to theory and experiment.

Note added in proof.—Similar conclusions were arrived at by Paglione *et al.* [30] on the basis of resistivity measurements in a wider field range. In addition, recent theoretical work [31] indicates that superconducting transition may indeed suppress antiferromagnetism.

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