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Avoided Antiferromagnetic Order and Quantum Critical Point in CeCoIn₅

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We measured specific heat and resistivity of heavy fermion CeCoIn₅ between the superconducting critical field $H_{c2} = 5T$ and 9 T, with field in the [001] direction, and at temperatures down to 50mK. At 5T the data show Non Fermi Liquid behavior down to the lowest temperatures. At field above 8T the data exhibit crossover from the Fermi liquid to a Non Fermi Liquid behavior. We analyzed the scaling properties of the specific heat, and compared both 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 (AF) in CeCoIn₅ with the quantum critical point in the vicinity of the H_{c2} . Below H_{c2} the AF phase which competes with the paramagnetic ground state is superseded by the superconducting transition.

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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.

Study of quantum critical points in heavy fermion systems has been a 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 states. The unconventional behavior near 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 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 $\alpha < 0$. Resistivity with an exponent less than two is also ubiquitous in these compounds.

Tuning the system through a QCP can be accomplished experimentally by varying sample's composition [2, 3], applying pressure [4], or applying magnetic field [5]. In non-stoichiometric 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 separate this origin of NFL behavior from the consequences of the proximity to a QCP. Hence the sto-

ichiometric compounds receive great deal of attention in the field of quantum criticality.

One class of such materials are Ce-based compounds, which have an antiferromagnetic (AF) ground state at ambient pressure. Hydrostatic pressure suppresses the magnetic ordering temperature T_N to zero at a critical pressure of the QCP. Such approach was used successfully for CeCu₂Ge₂ [8] CeRh₂Si₂ [9], CePd₂Si₂, CeIn₃ [4], and some other compounds. Alternatively, AF order can be suppressed by an applied magnetic field. When this was done in YbRh₂Si₂, 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 CeCoIn₅, which show that this compound has a QCP with 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 CeCoIn₅ is an ambient pressure superconductor. We argue that magnetic order which competes with the paramagnetic state is superseded by the superconducting state.

CeCoIn₅ is a tetragonal, quasi-2D compound, with layers of CeIn₃ separated by layers of CoIn₂. 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 CeCoIn₅ is unconventional, with lines of nodes in the energy gap, as demonstrated by specific heat and thermal conductivity [12] and NQR [13] measurements. Pauli limiting analysis [14] and thermal conductivity modulations in magnetic field [15] indicate that CeCoIn₅ is a singlet, $d_{x^2-y^2}$ superconductor. Recently it was shown that the superconducting transition in CeCoIn₅ in magnetic fields close to the upper critical field for [001] direction, $H_{c2} = 4.95T$, is first order below $T = 0.7$ K [15, 16]. NFL behavior of CeCoIn₅ at the field of 5 T persists over a large region of temperature [17], with $\gamma \propto -\log(T)$ between 0.4 K 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 Sommerfeld coefficient of

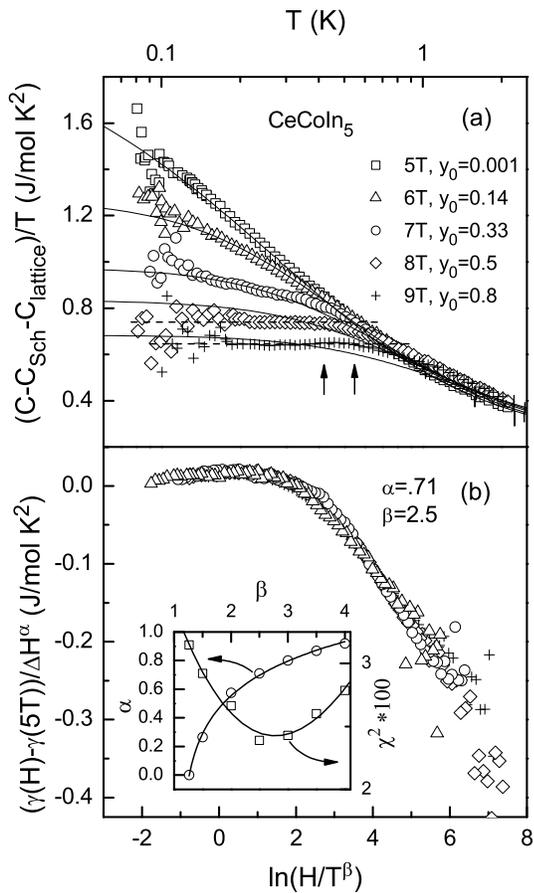


FIG. 1: (a) Sommerfeld coefficient, $\gamma(T) = C(T)/T$, of CeCoIn₅ in magnetic fields $H \parallel [001]$. Dashed lines for 8 T and 9 T emphasize the FL behavior with constant γ . Left (right) arrow indicate T_{FL}^C for 8 T (9 T). Solid lines are fits to the SCR spin-fluctuation model for each field 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 .

CeCoIn₅ versus temperature for several magnetic fields between 5 T and 9 T. Large low temperature Schottky anomaly tails due to the quadrupolar and magnetic spin splitting of In and Co nuclei were subtracted [12]. At high magnetic field and low temperature 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 studied. As the magnetic field is increased above 5 T, the low temperature data starts to deviate from the logarithmic behavior at ever higher temperature. Clear FL regime of $\gamma = \text{const.}$ at low temperature is recovered at 8 Tesla. Both 8 T 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 tempera-

ture range shown. This behavior is different from that of YbRh₂Si₂, where γ saturates below some temperature $T^*(H)$ to a constant value for all $H \neq H_c$, with a rather sharp break from the $-\log(T)$ behavior for $T > T^*$ [18].

The logarithmic divergence of $\gamma(T, H)$ at 5 T and γ 's increase followed by a crossover to the FL behavior at low T for higher fields is indicative of a field-tuned QCP near 5 T. 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 [19, 20].

In our case the tuning parameter is the applied magnetic field. Hence the energy scale of fluctuations away from the QCP depends on $|H - H_c|$. Therefore the entropy, and hence the specific heat, should scale as $\gamma(H) - \gamma(H_c) \propto f((H - H_c)/T^\beta)$. Experimentally, at fields sufficiently away from the QCP, we achieve the FL regime at low T . In that regime γ depends on the field but is T -independent. Consequently, the scaling relation has to take 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}Pd₃ [21], CeCu_{5.8}Ag_{0.8} [22], and for YbRh₂Si₂ [18].

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 CeCoIn₅ 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.

The crossover from the NFL to FL regimes is also clear from resistivity measurements in the same magnetic field range. Fig. 2(a) shows resistivity, ρ , of CeCoIn₅ below 5 K for fields between 5.6 T and 9 T, as well as the zero-field data. The data at 5.6 T and above is not affected by the proximity to the superconducting transition, as seen from the magnetoresistance measurements of CeCoIn₅ at 100 mK. [23] To focus on the low temperature behavior, we show the data below 1 K in 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 T 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

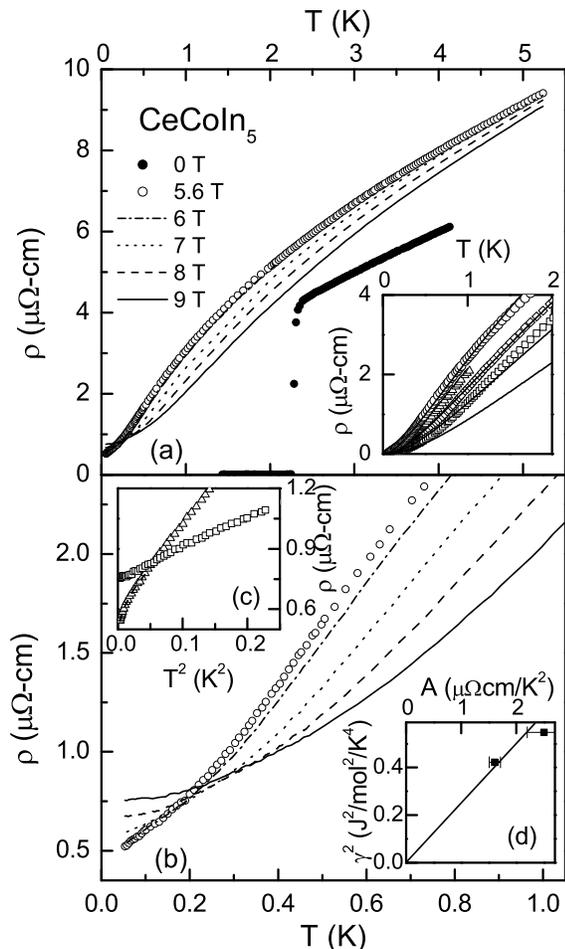


FIG. 2: (a) Resistivity of CeCoIn₅ in magnetic field $H \parallel [001]$ above the critical field $H_{c2} = 4.95$ T. Inset: Fits to $\rho(T)$ below 2K from the spin-fluctuation theory as detailed in the text. (c) 6 T (Δ), 7 T (\circ), 8 T (\diamond), 9 T (\square). Solid lines: $y_0 = 0.14, 0.33, 0.5,$ and 0.8 from top to bottom. (b) Low temperature region of data in (a). Inset (d): γ^2 vs. A . Solid line is a linear fit to the data points and the origin. Inset (c) Resistance for 6 T (Δ) and 9 T (\square) plotted vs. T^2 .

fields the linear in T , rather than quadratic, dependence of the resistivity is consistent with the NFL behavior of the specific heat. Qualitative difference in the temperature behavior of $\rho(T)$ at 6T and at 9T in the low temperature range is emphasized in the upper inset in Fig. 2(b), where resistivity is plotted versus T^2 for two fields, 9 T and 6 T. The 9 T data are linear in T^2 over the entire temperature range shown, while the 6 T data has a pronounced negative curvature.

To further analyze the FL region of the phase diagram of CeCoIn₅ in the inset (d) of Fig. 2(b) we plot γ^2 versus A for the 8 T and 9 T data. Within error bars the two values lie on the line $\gamma^2 = .3 \times A \text{ J}^2/\text{mol}^2\text{K}^2\mu\Omega\text{cm}$. The slope is about three times larger than the Kadowaki-Woods ratio [24], approximately obeyed by many heavy fermion compounds. This discrepancy is within the typ-

ical range of the scatter of the data for other HF compounds, and confirms the FL ground state of CeCoIn₅ for 8 T 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, CeRhIn₅, is an ambient pressure antiferromagnet with the Neel temperature $T_N = 3.8$ K, AF 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 AF spin fluctuations to the specific heat and scattering has been considered in Ref. 25 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 $2\pi^2T_0$ 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 CeCoIn₅ with CeRhIn₅. Pressure of 16 kbars suppresses the AF state in CeRhIn₅, at which point the superconducting state with $T_c = 2.1$ K emerges[26]. This lead Sidorov *et al.* to suggest that physical properties of CeCoIn₅ are very close to those of CeRhIn₅ at 16 kbars [27]. 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 CeRhIn₅. In the fit we assumed an additive field-independent contribution to $\gamma(T)$ of 0.2 J/mol K^2 from non-critical fermions. For the 5 T data the theoretical fits are identical for any $y_0 \leq 0.01$ over the temperature range studied. Consequently, CeCoIn₅ 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 AF order.

Finally, in Fig. 3 we plot the phase diagram of CeCoIn₅ in the $T - H$ plane. The normal-superconducting phase boundary was determined in Refs. 11, 16. At fields of 8T and 9T we see a clear transition from the NFL to FL behavior in both specific heat (T_{FL}^C) and resistivity (T_{FL}^p). Below 8T, in the temperature range studied, the

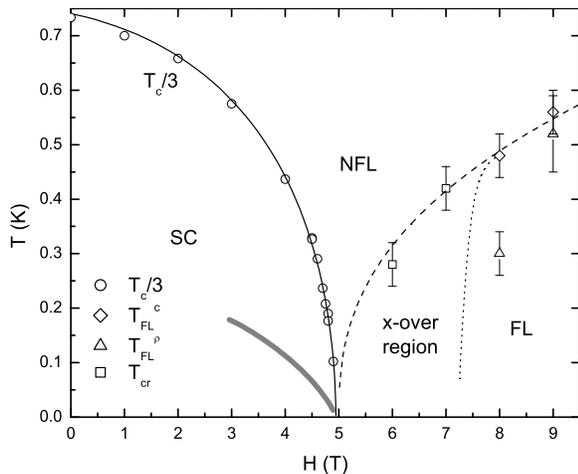


FIG. 3: Combined phase diagram of CeCoIn₅. Superconducting-normal phase boundary ((\circ) and solid line) is from Ref. 16. T_{cr} (\square) and the dashed line (guide to eye) denote the upper boundary of the crossover region. Dotted line through T_{FL}^c (\diamond) and T_{FL}^p (\triangle) schematically indicate the upper boundary of the FL region. Region between the dashed and dotted lines is the crossover region. Thick grey line is the hypothetical phase boundary of the AF state prevented by the superconducting transition.

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, while there is no signature of the AF ordered phase, the behavior of CeCoIn₅ above the superconducting upper critical field, $H_{c2} = 4.95\text{T}$, at low temperatures is controlled by a field-tuned antiferromagnetic quantum critical point. The existence of the QCP is confirmed by scaling analysis of the specific data. Incipient AF state in CeCoIn₅ is strongly suggested by the fits to both the specific heat and the resistivity based on the spin-fluctuation theory [25]. The underlying physical picture is that the AF 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 CeCoIn₅ 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 AF critical point. Our results imply that the transition temperature of the avoided antiferromagnetic order vanishes at fields very close to the superconducting upper critical field. This concomitant suppression of the AF and SC orders is unusual, and may reflect an important aspect of the underlying physics, presenting further challenges to theory and experiment.

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