Precursor State to Unconventional Superconductivity in CeIrIn_5

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We present Hall effect and magnetoresistance measurements in the heavy fermion superconductor CeIrIn_5. At low temperature, a Kondo coherent state is established. Deviations from Kohler's rule and a quadratic temperature dependence of the cotangent of the Hall angle are reminiscent of properties observed in the high-temperature superconducting cuprates. A striking observation pertains to the presence of a *precursor* state—characterized by a change in the Hall mobility—that precedes the superconductivity in this material, in similarity to the pseudogap in the cuprate superconductors.

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Heavy fermion superconductivity continues to be a central focus of investigations into strongly correlated electron systems. The initial interest was primarily centered on reconciling the observation of superconductivity in an inherently magnetic environment and its interplay with the Kondo screening effects in a correlated Fermi liquid. However, its reemergence has been dramatic, with current emphasis being placed on understanding the phenomenon of magnetic quantum critical points (QCPs). Here, a QCP refers to a zero temperature (T) magnetic instability that can be tuned by a nonthermal control parameter like the magnetic field (H), pressure (P), or chemical composition. Such a QCP is believed to crucially influence physical properties in a substantial region of the H-T-P phase space in its vicinity. The added incentive is to bridge our understanding of the heavy fermions and the high-temperature superconductors, two distinctly separate classes of systems where superconductivity and magnetism are intricately connected.

The more recent discovery of the $\mathrm{Ce}M\mathrm{In}_5$ (where M: Co, Rh, or Ir) family of heavy fermion systems has further enriched this field [1]. These layered materials crystallize in the tetragonal structure, and the quasi-two-dimensional character of their Fermi surfaces was confirmed by de Haas–van Alphen (dHvA) measurements [2]. Moreover, both the superconducting and normal states in these materials are highly unusual. For instance, in CeCoIn_5 several measurements indicated [3] that the superconducting gap function has line nodes and is most likely to have a d-wave symmetry. Coupled with other observations like a linear temperature dependent resistivity and a strongly temperature dependent Hall coefficient R_H, a remarkable similarity of these systems with the cuprate superconductors was suggested [4].

CeIrIn_5 is the other ambient pressure superconductor in this series, with a bulk $T_c \approx 0.4$ K and a resistive $T_c \approx 1.2$ K [5]. In spite of a band structure similar to its Co and Rh counterparts, striking differences have been observed in both its superconducting and normal state properties. The primary difference pertains to the position of the magnetic instability with respect to the superconducting region: In CeCoIn_5, the magnetic field tuned QCP appears to be close to the upper critical field $H_{c2}(T)$ of the superconductor [6–8]. However, in CeIrIn_5 the magnetic instability is reported to lie far away from the superconducting region as has been inferred from prior investigations [9,10] with the applied field suppressing rather than enhancing the Landau Fermi liquid (FL) state. This has also led to suggestions that CeIrIn_5 may be a prospective system, in which superconductivity is mediated by charge (valence) fluctuations [11]. Importantly, unlike its Co counterpart, the H-T phase space in the vicinity of the superconducting region in CeIrIn_5 is expected to be free from the influence of the magnetic instability, thus enabling a cleaner investigation of the superconducting state. In this Letter, we report the investigation of CeIrIn_5 using sensitive magnetoresistance and Hall effect measurements. Besides the observation of a low-temperature Kondo state, experimental signatures of the presence of a *precursor* state that envelops the superconducting region in CeIrIn_5 is seen—an observation that could imply that the formation of the superconducting condensate is preceded by an electronic state hitherto unexplored in this class of materials.

The magnetotransport measurements were conducted as isothermal field sweeps on high quality single crystals of CeIrIn_5 (resistivity $\rho \approx 1.75\ \mu\Omega\,\mathrm{cm}$ at 1.35 K), with the crystallographic c axis parallel to the applied field and a current of 20 μA being applied within the ab plane. In addition, temperature sweeps were carried out at selected fields to complement the typically more sensitive isothermal measurements. The setup is based on [8], with additional low-noise preamplifiers to enhance the sensitivity to about ± 0.01 nV. The Hall voltage is obtained as the asymmetric component under field reversal.

The magnetoresistance $\mathrm{MR} = [\rho_{xx}(H) - \rho_{xx}(0)]/\rho_{xx}(0) = \Delta\rho_{xx}/\rho_{xx}(0)$ as measured in CeIrIn_5 at selected temperatures is shown in Fig. 1. At the lowest measured temperatures, the MR is positive (due to the Lorentz force)

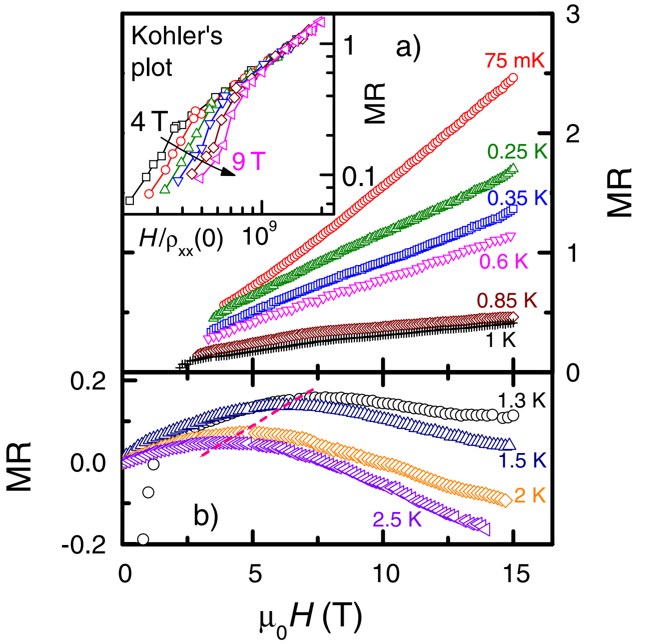

FIG. 1 (color online). Magnetoresistance, $\mathrm{MR} = \Delta\rho_{xx}(H)/\rho_{xx}(0)$, as measured in CeIrIn$_5$ at constant $T \leq 1$ K (a) and $T > 1$ K (b). The maximum in MR indicates the onset of Kondo coherence, dashed line in (b). Inset to (a): Violation of Kohler's scaling rule in the NFL regime. In this plot, T is an implicit parameter, and the abscissa is given in units of T/Ω m.

and almost linear as a function of applied field. With increasing temperature, a negative contribution to the MR is seen to arise at high fields, with competition between the negative and positive contributions eventually resulting in a *crossover,* where the sign of $\partial(\mathrm{MR})/\partial H|_T$ changes [dashed line in Fig. 1(b)]. The negative component of MR stems from the suppression of spin flip scattering. Hence, with increasing temperature it is expected to grow and, consequently, the crossover should move to lower fields, as is indeed observed in our data. This crossover is usually referred to as the *onset* of the Kondo coherent regime at $H_{\mathrm{coh}}(T)$ [12]. Our results are in agreement with those of prior optical conductivity measurements that indicate a low-temperature coherent state in CeIrIn$_5$ [13]. A recent two fluid description of the Kondo lattice suggested that the $T \rightarrow 0$ ground state in these materials can be described by a sum of a (single Ce^{3+} ion) Kondo gas and a coherent Kondo liquid, with the latter being about 95% of the whole in the case of CeIrIn$_5$ [14]. This Kondo coherence is expected to form below $T \approx 20$ K [14]. In the low-temperature limit, this coherence scale of the Kondo lattice is anticipated to vanish near the magnetic instability ($\gtrsim 25$ T [15]).

In the FL description, positive MR arises from the bending of the charge carrier trajectory by the Lorentz force. Assuming isotropic scattering times at all points on the Fermi surface, the MR is expected to scale as a function of $H/\rho_{xx}(0)$. This is known as Kohler's rule [16], and should hold regardless of the topology and the symmetry of the Fermi surface. The inset of Fig. 1(a) exhibits a Kohler's plot for CeIrIn$_5$, clearly indicating a violation of Kohler's rule. The deviation from scaling occurs in the non-Fermi liquid (NFL) regime, and the temperature and field dependences of this transition match with prior observations [9].

The Hall effect can be of great significance in the investigation of heavy fermion systems in the vicinity of a QCP [17]. In these systems, the low-temperature Hall coefficient predominantly arises from the normal part of the Hall effect [18], and thus reflects the evolution of the Fermi surface volume. The results for the Hall resistivities $\rho_{xy}(H)$ in CeIrIn$_5$ at selected temperatures are shown in Fig. 2. The measured ρ_{xy} are negative (indicating electron-dominated transport) and nonlinear in field down to the lowest measured temperature (50 mK). Their magnitudes are in good agreement with prior high-temperature data [19]. At low temperatures, $\rho_{xy}(H)$ exhibits a nearly quadratic field dependence. This quadratic regime is valid only for higher fields as the temperature is increased. Interestingly, in spite of the complex (renormalized) band structure of CeIrIn$_5$, the observed $\rho_{xy}(H)$ behavior can—at least qualitatively—be explained on the basis of that expected for simple compensated metals. Such a quadratic field dependence is anticipated [20] in the high field limit, i.e., when $\omega_c\tau \gg 1$ ($\omega_c = eH/m^*$ is the cyclotron frequency, m^* is the effective mass, and τ is the average time between scattering events). Since CeIrIn$_5$ *is* a compensated metal, as concluded from dHvA measurements [21], this observed field dependence is reasonable. A decrease in τ with increasing temperatures explains the shift of the quadratic regime to higher fields at high temperatures (Fig. 2). Here we note that in spite of the reported similar band

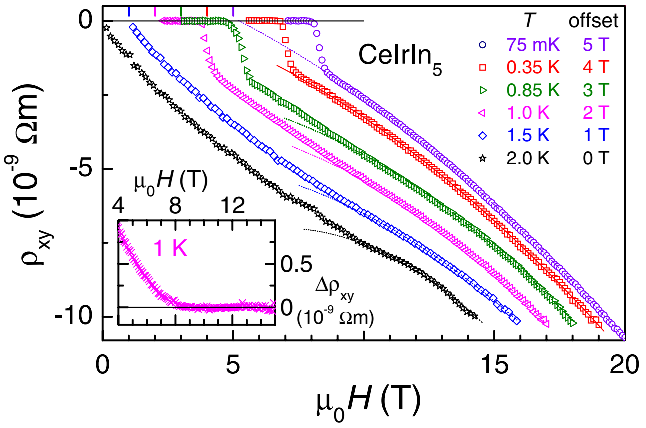

FIG. 2 (color online). Isothermal Hall resistivity $\rho_{xy}(H)$. For clarity, each data set is offset along the magnetic-field axis (offset value given in the legend). The sharp drop of ρ_{xy} to zero for $T < 1.2$ K indicates the onset of superconductivity. The dotted lines indicate the regime of H^2 dependence that shifts to higher fields with increasing temperature. Inset: Difference $\Delta(\rho_{xy})$ between measured values ρ_{xy} and the quadratic fit at $T = 1$ K.

structure of the Ir and Co based 115 systems there are obvious differences in their Hall resistivities: In CeCoIn$_5$, $\rho_{xy}(H)$ was linear at the lowest temperature and a (P dependent) signature in the Hall coefficient R_H was observed presumably resulting from critical spin fluctuations [8]. A likely reason for this behavior *not* being observed in CeIrIn$_5$ could be that the field range explored by our measurements does not encompass a putative QCP, a consequence of the fact that the magnetic instability in each system lies at very different field values.

The cotangent of the Hall angle ($\cot\theta_H = \rho_{xx}/\rho_{xy}$) is directly related to the charge carrier mobility and is a quantity of fundamental interest [22]. In many systems, including the cuprates, it has been observed to vary as T^2. Since ρ_{xx} in cuprates is observed to be linear in temperature, this functional form of $\cot\theta_H$ reflects a Hall scattering rate (τ_H^{-1}), which is at variance with the scattering rate ($\tau_{\rm tr}^{-1}$) governing the resistivity. Figure 3(a) exhibits the T^2 dependence of $\cot\theta_H$ as deduced in CeIrIn$_5$, a behavior observed in a substantial region of the H-T phase space. Interestingly, however, systematic deviations are seen at low temperatures. Though this aspect has rarely been addressed in the context of heavy fermion systems, such deviations from T^2 have been used to mark the onset of the pseudogap phase in some cuprate superconductors [23]. Our measurement protocol enables us to evaluate in more detail the field dependence of this quantity, and careful inspection of the H-T phase space in the vicinity of the superconducting region shows that $\cot\theta_H$ has a H^{-1} dependence. Interestingly, as one decreases temperature and approaches the superconducting region, systematic deviations from this H^{-1} dependence are observed below a critical field H^*. This is shown for the example of $T = 1$ K in Fig. 3(b). The difference $\Delta(\cot\theta_H)$ of the experimental data $\cot\theta_H$ from a linear fit [Fig. 3(c)] is used to identify H^*. We emphasize that this deviation is also reflected as a subtle feature in the field dependence of the MR [Fig. 3(d)].

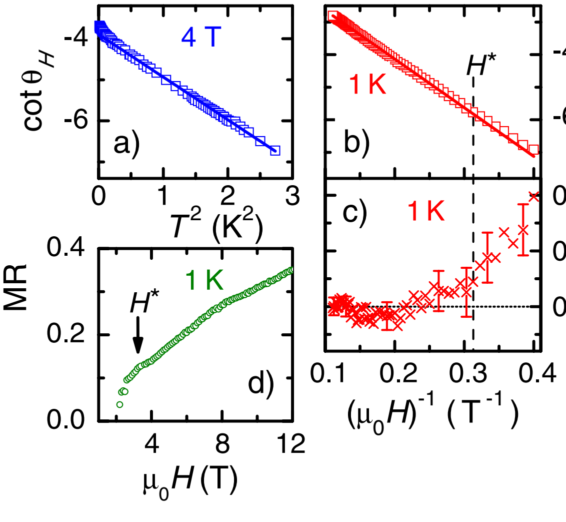

FIG. 3 (color online). (a) T^2 dependence of $\cot\theta_H$. (b) Dependence of $\cot\theta_H$ on H^{-1} at $T = 1$ K. Experimental data $\cot\theta_H$ (□) deviate from a linear H^{-1} dependence (line) below H^*. (c) Difference $\Delta(\cot\theta_H)$ between $\cot\theta_H$ and a linear fit to $\cot\theta_H$. H^* is identified by this difference exceeding the error of $\cot\theta_H$. (d) H dependence of the MR at 1 K. The anomaly at H^* is clearly visible.

Attempts to reconcile the observed functional form of $\cot\theta_H$ in cuprates with theory have primarily been based on (i) a model within the Luttinger liquid formalism, which relates the different scattering rates to distinct quasiparticles with dissimilar scattering events [24] and (ii) a nearly antiferromagnetic FL description, which predicts anisotropic scattering on the Fermi surface, with τ_H^{-1} and $\tau_{\rm tr}^{-1}$ being dictated by scattering events on different parts of the Fermi surface [25]. In Anderson's theory [24], the Hall angle is governed only by τ_H^{-1} and can be expressed as $\cot\theta_H = 1/\omega_c\tau_H$. Thus, $\cot\theta_H$ would be expected to vary as H^{-1} (with the slope being a function of τ_H^{-1}) as is observed in our case. Figure 4 shows the H-T phase diagram of CeIrIn$_5$, with the FL-NFL transition as determined from the deviations from Kohler's rule (inset of Fig. 1), the onset of Kondo coherence below $H_{\rm coh}$ determined from the maximum in MR [Fig. 1(b)], and the deviations from H^{-1} at H^* [Fig. 3(b)] clearly marked out. The most striking feature here is the envelope of H^* around the superconducting region indicating that the formation of a superconducting condensate in CeIrIn$_5$ is preceded by a *precursor* state associated with a change in the Hall mobility. Moreover, the critical field $H^*(T)$ of this precursor state can be scaled onto $H_{c2}(T)$ (inset to Fig. 4), suggesting that both these states might arise from the same underlying mechanism.

For the system CeCoIn$_5$, a precursor state has also been deduced from thermopower and Nernst effect measure-

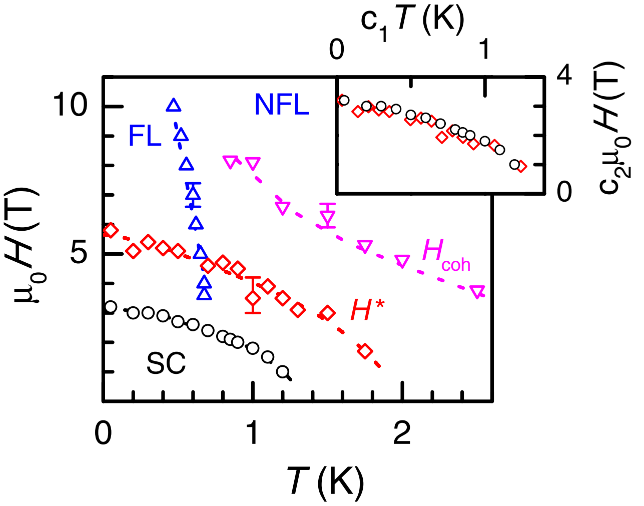

FIG. 4 (color online). H-T phase diagram of CeIrIn$_5$ determined from a combination of Hall effect and MR measurements (lines are guides to the eye). Representative error bars are given. Inset: Scaling of $H^*(T)$ with $H_{c2}(T)$, with $c_1 = c_2 = 1$ for $H_{c2}(T)$ and $c_1 = 0.7$, $c_2 = 0.55$ for $H^*(T)$.

ments [26]. In analogy with the cuprates, a vortex-liquid state was suggested, where thermal phase and vortex fluctuations result in short-range phase coherence [27]. Though this cannot be ruled out as the cause of our experimental observations, we note that we have failed to observe a measurable Hall signal in the Shubnikov state of $\mathrm{CeIrIn_5}$, probably indicating that vortex dynamics is rather weak. Moreover, our prior investigations failed to reveal a diamagnetic response in this phase space region.

An alternative scenario involves a strong anisotropy of the transport scattering rates, which in turn arises from the coupling of antiferromagnetic fluctuations to the (otherwise isotropic) FL formalism. This is achieved by the formation of hot (and cold) spots on different regions of the Fermi surface. Here, hot spots refer to positions on the Fermi surface, where the antiferromagnetic Brillouin zone intersects it, and the electron lifetimes (τ_{hot}) are very short. Thus, all the transport coefficients would be renormalized with respect to the ratio $\tau_{\mathrm{cold}}/\tau_{\mathrm{hot}}$, reflecting the anisotropy of the Fermi surface. An increasing magnetic field is expected to suppress these antiferromagnetic fluctuations, thus effectively *closing* the gapped regions of the Fermi surface. It is to be noted that transport [28] and nuclear quadrupole resonance (NQR) measurements [29] have been used to speculate on the presence of a pseudogap phase in the Co and Rh counterparts, respectively. A related scenario was recently reported: an *anisotropic* destruction of the Fermi surface in $\mathrm{CeCoIn_5}$ in the $T \to 0$ limit, reminiscent of the pseudogap phase in the cuprates [30]. A similar mechanism could be at play in $\mathrm{CeIrIn_5}$ considering the presence of anisotropic spin fluctuations in this system as inferred from NQR measurements [31].

At present, one can only speculate on the nature of low-lying electronic excitations responsible for this precursor state. For instance, it may arise as a consequence of antiferromagnetic fluctuations or may even signify a hitherto unknown form of unconventional correlations. The scaling of $H^*(T)$ with $H_{c2}(T)$ points towards a common origin of the precursor state and the superconductivity in this system. The FL-NFL crossover in the phase diagram is related to the presence of the magnetic instability at $\mu_0 H \approx 25$ T. This instability is also expected to influence $H_{\mathrm{coh}}(T)$, and a crossing between the FL and $H_{\mathrm{coh}}(T)$ lines is improbable. Moreover, the precursor state encompasses *both* the FL and the NFL regimes. This is in contrast to observations in the cuprates implying that theoretical approaches commonly employed in the latter may have only limited applicability here. The low-temperature phase diagram of $\mathrm{CeIrIn_5}$ is clearly influenced by both the magnetic instability and the presence of the precursor state. Whether they *complement* or *compete* with each other, is an aspect that more direct experiments would need to resolve.

In summary, Hall effect and MR measurements clearly demarcate the low-temperature Kondo coherent state and the FL-NFL transition of $\mathrm{CeIrIn_5}$. The most striking observation, however, is the presence of a pseudogap-type precursor state preceding superconductivity in this system, which is characterized by a change in the Hall mobility. A microscopic comprehension of this precursor state would be crucial, not only for understanding the electron pairing in this system, but also in placing it in proper perspective to the superconducting cuprates.

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