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A precursor state to unconventional superconductivity in CeIrIn₅

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We present sensitive measurements of the Hall effect and magnetoresistance in CeIrIn₅ down to temperatures of 50 mK and magnetic fields up to 15 T. The presence of a low temperature coherent Kondo 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. The most striking observation pertains to the presence of a *precursor* state—characterized by a change in the Hall mobility—that appears to precede the superconductivity in this material, in similarity to the pseudogap in the cuprate high T_c superconductors.

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

The more recent discovery of the CeMIn₅ (where M : Co, Rh or Ir) family of HF 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 (FS) was confirmed by de Haas-van Alphen (dHvA) measurements [2]. Moreover, both the superconducting and normal states in these materials have been reported to be highly unusual. For instance, in CeCoIn₅ (with the highest reported ambient pressure superconducting transition temperature $T_c \approx 2.3$ K among Ce-based HF systems [1]) measurements of specific heat, thermal conductivity and Andreev reflection have 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 T dependent resistivity and a strongly T dependent Hall coefficient R_H , a remarkable similarity of these systems with the high T_c cuprates was suggested [4].

CeIrIn₅ 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 SC region in these systems. In CeCoIn₅, the magnetic field tuned QCP appears to be close to the upper critical field $H_{c2}(T)$ of the superconductor [6, 7, 8]. However, in CeIrIn₅ the magnetic instability is reported to lie far away from the superconducting region as has been inferred from prior investigations using H , P and chemical composition as control parameters; with H suppressing rather than enhancing the Landau Fermi liquid (FL) state [9, 10]. This has also led to suggestions that CeIrIn₅ may be a prospective system, in which SC is mediated by charge valence fluctuations [11]. Unlike its Co counterpart, the H - T phase space in the vicinity of the SC region in CeIrIn₅ is expected to be free from the influence of the magnetic instability, thus enabling a cleaner investigation of the superconducting state in this HF system. In this letter, we report the investigation of CeIrIn₅ using sensitive magnetoresistance and Hall effect measurements. Besides the observation of a low-temperature coherent Kondo state, experimental signatures of the presence of a *precursor* state that envelops the superconducting region in this system is seen—an observation which could imply that the condensation of electrons into Cooper pairs is preceded by an electronic state hitherto unexplored in this class of materials.

The magnetotransport measurements were mainly conducted as isothermal field sweeps on high quality single crystals of CeIrIn₅ (resistivity $\rho \approx 1.75 \mu\Omega$ at 1.35 K), with the crystallographic c axis parallel to H and the current of $\approx 20 \mu\text{A}$ being applied along the ab plane. In addition, temperature sweeps were carried out at selected H to complement the typically more sensitive isothermal measurements. The set up is based on [8], with additional low-noise preamplifiers used to enable a sensitivity of the order of ± 0.01 nV. The Hall voltage is obtained as

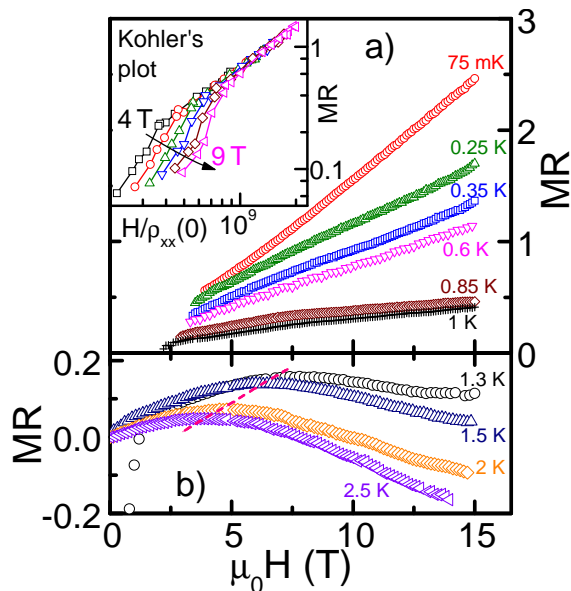


FIG. 1: Magnetoresistance, $MR = \Delta\rho_{xx}/\rho_{xx}(0)$, as measured in CeIrIn₅ at constant $T \leq 1$ K (a) and $T > 1$ K (b). The maximum in MR indicates the coherent transition, dashed line in (b). Inset to (a): Violation of Kohler's scaling rule in the NFL regime. In this plot, T is an implicit parameter, and the abscissa is given in units of $T/\Omega m$.

the asymmetric component under field reversal.

The magnetoresistance $MR = [\rho_{xx}(H) - \rho_{xx}(0)]/\rho_{xx}(0) = \Delta\rho_{xx}/\rho_{xx}(0)$ as measured in CeIrIn₅ at selected temperatures is shown in Fig. 1. At the lowest measured T , the MR is seen to be positive and subquadratic as a function of H . With increasing T , a negative contribution to the MR is seen to arise at high H , with competition between the negative and positive contributions eventually resulting in a *crossover*, where the sign of $\partial(MR)/\partial H|_T$ changes. This crossover can be identified to be the *coherent* transition [12], marking the onset of the dense Kondo regime at H_{coh} [dashed line in Fig. 1(b)]. Since the negative component of MR stems from the suppression of spin flip scattering, it is expected to grow with increasing T . This should result in the crossover moving to lower H with increasing T , as is indeed observed in our data. These measurements are in agreement with prior optical conductivity measurements which indicate the formation of a low- T coherent state in CeIrIn₅ [13]. A recent two fluid description of the Kondo lattice has suggested that the $T \rightarrow 0$ ground state in these materials can be described by a sum of the (single Ce³⁺ ion) Kondo gas and a coherent Kondo liquid, with the latter being about 95% of the whole in the case of CeIrIn₅ [14]. In the high- T limit, this Kondo coherence would be expected to form below $T \approx 20$ K [14]. In the low- T limit, this coherence scale of the Kondo lattice is anticipated to vanish at the magnetic instability ($\gtrsim 25$ T, [15, 16]).

In the FL description, the low- T positive MR arises

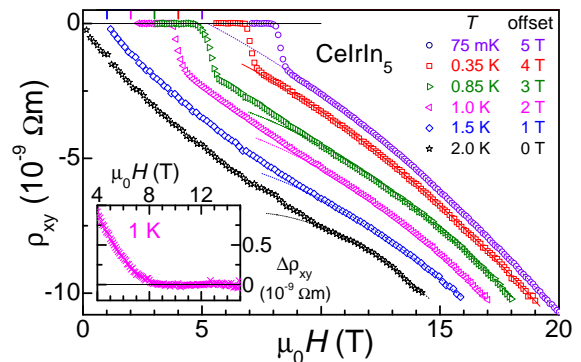


FIG. 2: Isothermal Hall resistivity $\rho_{xy}(H)$. For clarity, each data set is offset along the H axis, with the offset value indicated in the legend. The dotted lines (guides to the eye) indicate the quadratic H regime which shifts to higher fields with increasing T . Inset: The difference $\Delta(\rho_{xy})$ between measured values ρ_{xy} and the quadratic fit, for $T = 1$ K.

from the bending of the electron trajectory by the Lorentz force. Assuming isotropic scattering times at all points on the FS, the MR is expected to scale as a function of $H/\rho_{xx}(0)$. This is known as Kohler's rule [17], and should hold regardless of the topology and the symmetry of the FS. The inset of Fig. 1(a) exhibits a Kohler's plot for CeIrIn₅, clearly indicating a violation of Kohler's rule. The deviation from scaling occurs in the non Fermi liquid (NFL) regime, and the T and H dependences of this transition match well with prior reports [9].

The Hall effect, a rather complex quantity, has proven to be of great significance in the investigation of HF systems in the vicinity of a QCP [18]. This is due to the fact that in HF systems, the low temperature R_H predominantly arises from the normal part of the Hall effect [19], and thus can be used to monitor the evolution of the FS volume. The results for the Hall resistivities $\rho_{xy}(H)$ in CeIrIn₅ at selected T are shown in Fig. 2. The measured ρ_{xy} is seen to be negative (indicating electron-dominated transport) and nonlinear in H down to the lowest measured T . Their magnitudes are in good agreement with prior high-temperature data [20]. At low T , ρ_{xy} exhibits a nearly quadratic H dependence, with this quadratic regime only valid for higher fields as T is increased. It is interesting to note that in spite of the complex band structure of CeIrIn₅, the observed ρ_{xy} behavior can—at least qualitatively—be explained on the basis of that expected for simple compensated metals. Here, a quadratic H dependence is anticipated [21] 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 the average time between scattering events). Since CeIrIn₅ is a compensated metal, as concluded from dHvA measurements [22], this observed H dependence is not unexpected. A decrease in τ with increasing T explains the shift of the quadratic regime to higher fields at higher T . In this con-

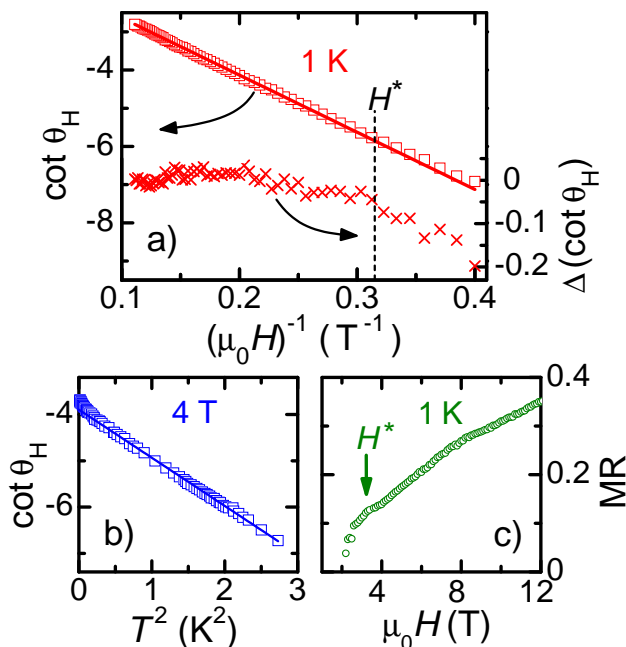


FIG. 3: (a) H^{-1} dependence of $\cot \theta_H$ at $T = 1$ K. Experimental data $\cot \theta_H$ deviate from a linear H^{-1} dependence below H^* . The difference $\Delta(\cot \theta_H)$ (\times and right scale) between $\cot \theta_H$ and a linear fit to $\cot \theta_H$ is used to identify H^* . (b) T^2 dependence of $\cot \theta_H$. (c) H dependence of the MR at 1 K, with the anomaly at H^* marked by an arrow.

text we note that in spite of the reported similarity of the Ir and Co based systems with respect to their band structure, there are obvious differences in ρ_{xy} as measured in CeIrIn_5 with that of its Co counterpart reported earlier [8]. For instance, in CeCoIn_5 ρ_{xy} was linear at the lowest T and a (P dependent) signature in R_H was observed which was attributed to arise as a consequence of critical spin fluctuations. A likely reason for this behavior *not* being observed in CeIrIn_5 could be that the H - T phase space explored by our measurements does not encompass the putative QCP, a consequence of the fact that the magnetic instability in each system lies in very different regions of the H - T phase space.

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 [23]. 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 T , this functional form of $\cot \theta_H$ reflects a Hall scattering rate (τ_H^{-1}) which is at variance with the scattering rate (τ_{tr}^{-1}) governing the resistivity. Fig. 3(b) 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 T . Though this aspect has not been addressed in the context of HF systems, such deviations from T^2 have been used to mark the onset of the pseudogap phase

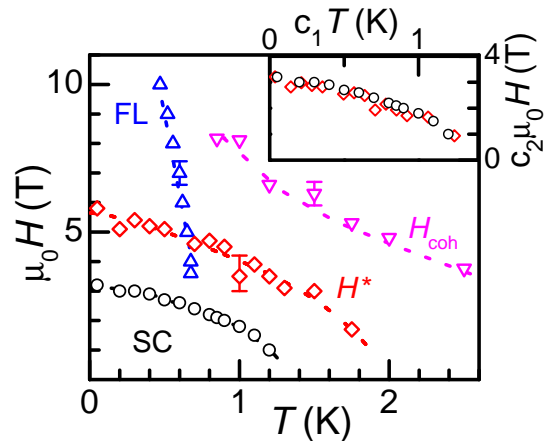


FIG. 4: H - T phase diagram of CeIrIn_5 determined from a combination of Hall effect and MR measurements (lines are guides to the eye). Inset: Scaling of $H^*(T)$ with $H_{c2}(T)$, with $c_1 = c_2 = 1$ for $H_{c2}(T)$ and $c_1 = 0.7$, $c_2 = 0.55$ for $H^*(T)$.

in some high T_c cuprates [24]. Our measurement protocol enables us to evaluate in more detail the H dependence of this quantity, and careful inspection of the H - T phase space in the vicinity of the SC region shows that $\cot \theta_H$ has a H^{-1} dependence. Interestingly, as one decreases T 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(a). The difference $\Delta(\cot \theta_H)$ of the experimental data $\cot \theta_H$ from a linear fit (\times and right axis) is used to identify H^* below which $\cot \theta_H$ deviates from H^{-1} . We emphasize that this deviation is also reflected as subtle feature in the H dependence of the MR, see Fig. 3(c).

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 particles with dissimilar scattering events [25] and (ii) a nearly antiferromagnetic (AF) FL description, which predicts anisotropic scattering on the FS, with τ_H^{-1} and τ_{tr}^{-1} being dictated by scattering events on different parts of the FS [26]. In Anderson's theory [25], 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. Fig. 4 shows the H - T phase diagram of CeIrIn_5 , with the FL-NFL transition as determined from deviations from Kohler's rule (inset of Fig. 1), the onset of Kondo coherence determined from the maximum in MR, Fig. 1(b), and the deviations from H^{-1} at H^* [Fig. 3(a)] clearly marked out. The most striking feature here is the envelope of H^* around the superconducting region, indicating that the condensation of itinerant electrons into Cooper pairs 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 $H_{c2}(T)$ (as shown in the inset) suggesting that both these states might arise from the same underlying mechanism.

For the system CeCoIn₅, a precursor state has also been deduced from thermopower and Nernst effect measurements [27]. In analogy with the cuprates, a vortex-liquid state was suggested, where thermal phase and vortex fluctuations result in short-range phase coherence [28]. 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 mixed state of CeIrIn₅, probably indicating that vortex dynamics is rather weak. Moreover, prior investigations have failed to reveal a diamagnetic response in this phase space region.

An alternative scenario would involve a strong anisotropy of the transport scattering rates, which in turn arise from the coupling of AF fluctuations to the (otherwise isotropic) FL formalism. This is achieved by the formation of hot (and cold) spots on different regions of the FS. Here, hot spots refer to positions on the FS surface, where the AF Brillouin zone intersects it, and the electron lifetimes are very short. Thus, all the transport coefficients would be renormalized with respect to the ratio τ_{cold}/τ_{hot} , reflecting the anisotropy of the FS. An increasing H would be expected to suppress these AF fluctuations, thus effectively *closing* the gapped regions of the FS. It is to be noted that transport [29] and (P dependent) nuclear quadrupole resonance (NQR) measurements [30] 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 FS in CeCoIn₅ in the $T \rightarrow 0$ limit, reminiscent of the pseudogap phase in the cuprates [31]. In spite of the absence of a magnetic instability in the immediate vicinity of the SC region in CeIrIn₅ (in contrast to its Co counterpart), a prior NQR study has inferred on the presence of anisotropic spin fluctuations in CeIrIn₅ [32] indicating that a similar mechanism could be at play in this system.

In the absence of other experimental evidences, one can only speculate on the nature of low-lying electronic excitations which give rise to this precursor state. It may arise as a consequence of AF fluctuations as discussed above, or may even signify a hitherto unknown form of unconventional order. The scaling of $H^*(T)$ with $H_{c2}(T)$ is striking, and points towards a common origin of the precursor state and the SC 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 would also be expected to influence H_{coh} , and a crossing between the FL and H_{coh} lines is improbable. However, it is pertinent to note that the precursor state encompasses both the FL and NFL regimes, and is suppressed to $T \rightarrow 0$ by the applied H in the FL regime of the phase diagram. This is in contrast to what

is observed in the cuprates implying that theoretical approaches commonly employed in the latter may have only limited applicability in this case. The low- T phase diagram of CeIrIn₅ is clearly dictated by both, the magnetic instability as well as the presence of the precursor state. Whether they *complement* or *compete* with each other, is an aspect which more direct experiments would need to resolve.

In summary, Hall effect and MR measurements clearly demarcate the low- T coherent Kondo state and the FL–NFL transition of CeIrIn₅. The most striking observation, however, is the presence of a pseudogap-type precursor state preceding the SC 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 with respect to the high T_c cuprates.

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- [1] C. Petrovic *et al.*, J. Phys.: Condens. Matter **13**, L337 (2001).
 - [2] R. Settai *et al.*, J. Phys.: Condens. Matter **13**, L627 (2001).
 - [3] Y. Matsuda *et al.*, J. Phys.: Condens. Matter **13**, R705 (2006).
 - [4] Y. Nakajima *et al.*, J. Phys. Soc. Jpn. **76**, 024703 (2007).
 - [5] C. Petrovic *et al.*, Europhys. Lett. **53**, 354 (2001).
 - [6] J. Paglione *et al.*, Phys. Rev. Lett. **91**, 246405 (2003).
 - [7] A. Bianchi *et al.*, Phys. Rev. Lett. **91**, 257001 (2003).
 - [8] S. Singh *et al.*, Phys. Rev. Lett. **98**, 57001 (2007).
 - [9] C. Capan *et al.*, Phys. Rev. B. **70**, 180502(R) (2004).
 - [10] M. Nicklas *et al.*, Phys. Rev. B **70**, 020505(R) (2004).
 - [11] A. T. Holmes *et al.*, J. Phys. Soc. Jpn. **76**, 051002 (2007).
 - [12] N. B. Brandt and V. V. Moshchalkov, Adv. Phys. **33**, 373 (1984).
 - [13] F. P. Mena, D. van der Marel, and J. L. Sarrao, Phys. Rev. B **72**, 045119 (2005).
 - [14] S. Nakatsuji, D. Pines, and Z. Fisk, Phys. Rev. Lett. **92**, 016401 (2004).
 - [15] T. Takeuchi *et al.*, J. Phys. Soc. Jpn. **70**, 877 (2001).
 - [16] J. S. Kim *et al.*, Phys. Rev. B **65**, 174520 (2002).
 - [17] A. B. Pippard, *Magnetoresistance in Metals* (Cambridge University Press, Cambridge 1989).
 - [18] S. Paschen *et al.*, Nature (London) **432**, 881 (2004).
 - [19] A. Fert and P. M. Levy, Phys. Rev. B **36**, 1907 (1987).
 - [20] M. F. Hundley, Phys. Rev. B **70**, 035113 (2004).
 - [21] C. M. Hurd, *The Hall effect of metals and alloys* (Plenum Press, New York, 1972).
 - [22] Y. Haga *et al.*, Phys. Rev. B **63**, 060503(R) (2001).
 - [23] T. R. Chien *et al.*, Phys. Rev. Lett. **67**, 2088 (1991).

- [24] Y. Abe *et al.*, Phys. Rev. B **60**, R15055 (1999).
- [25] P. W. Anderson, Phys. Rev. Lett. **67**, 2092 (1991).
- [26] B. P. Stojkovic, D. Pines, Phys. Rev. B **55**, 8576 (1997).
- [27] R. Bel *et al.*, Phys. Rev. Lett. **92**, 217002 (2004).
- [28] Y. Onose *et al.*, Europhys. Lett. **79**, 17006 (2007).
- [29] V. A. Sidorov *et al.*, Phys. Rev. Lett. **89**, 157004 (2002).
- [30] S. Kawasaki *et al.*, Phys. Rev. B **65**, 020504(R) (2001).
- [31] M. A. Tanatar *et al.*, Science **316**, 1320 (2007).
- [32] G.-q. Zheng *et al.*, Phys. Rev. Lett. **86**, 4664 (2001).