

Anomalous magnetotransport in the heavy-fermion superconductor Ce_2PdIn_8

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The normal-state behavior in the heavy-fermion superconductor Ce_2PdIn_8 has been probed by means of Hall coefficient (R_H) and transverse magnetoresistivity (MR) measurements. The results indicate the predominance of contributions from antiferromagnetic spin fluctuations at low temperatures. Anomalous non-Fermi-liquid-like features, observed below 8 K in both $R_H(T)$ and $\text{MR}(T)$, are related to the underlying quantum critical point, evidenced before in the specific heat and the electrical resistivity data. The magnetotransport in Ce_2PdIn_8 is shown to exhibit specific types of scaling that may appear universal for similar systems at the verge of magnetic instability.

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In the vicinity of the quantum critical point (QCP), which separates an ordered phase from a disordered phase at zero temperature, the finite-temperature physical properties of heavy-fermion (HF) systems are notably different from those predicted by the standard Fermi liquid (FL) theory of metals. As far as the magnetotransport properties are concerned, quantum criticality in two-dimensional (2D) antiferromagnetic (AF) systems manifests itself as linear-in- T electrical resistivity (ρ), strongly temperature- and field-dependent Hall coefficient (R_H), and the magnetoresistivity (MR) obeying the so-called modified Kohler's rule.¹

There is still a growing number of HF superconductors since the seminal discovery of unconventional superconductivity in CeCu_2Si_2 .² Recently, a compound Ce_2PdIn_8 has been identified as an ambient-pressure HF superconductor with a strongly enhanced normal-state electronic specific heat coefficient $\gamma \simeq 1.2 \text{ J}/(\text{mol K}^2)$ and the transition temperature $T_c \simeq 0.7 \text{ K}$.³⁻⁵ The superconductivity in this material has been found to possess a nodal character,⁶ likely of d -wave type, doubtlessly established for the closely related phase CeCoIn_5 .⁷ Furthermore, Ce_2PdIn_8 has been found to exhibit a magnetic-field-induced QCP near the upper critical field $H_{c2}(0) = 2.4 \text{ T}$,^{6,8,9} again in strong similarity to CeCoIn_5 .¹⁰ Most intriguingly, it has also been suggested that Ce_2PdIn_8 may fulfill the necessary conditions for the formation of a unique modulated Fulde-Ferrell-Larkin-Ovchinnikov superconducting state,⁶ possible occurrence of which at the magnetic high-field-low-temperature corner of the H - T phase diagram of CeCoIn_5 is presently being hotly debated.¹¹ The many common features between CeCoIn_5 and Ce_2PdIn_8 motivate further comprehensive studies of the physical behavior in the latter compound.

In this Rapid Communication, we report on the Hall effect and the transverse magnetoresistivity of Ce_2PdIn_8 , which both appeared highly anomalous yet consistent with the quantum criticality scenario, previously evoked based on the electrical resistivity,^{5,6,8} thermoelectric,¹² and heat capacity^{4,9} data. The polycrystalline sample used in the present study was previously characterized by means of x-ray diffraction, magnetic susceptibility, electrical resistivity, and heat capacity measurements.⁵ Detailed metallurgical examination, in concert with the obtained physical property data, indicated a single phase material, in particular one free of any contamination by

CeIn_3 impurity.⁴ The magnetotransport measurements were performed on a thin plateletlike polycrystalline specimen with surface dimensions of $2.5 \times 1.5 \text{ mm}$ and a thickness of $25 \pm 5 \mu\text{m}$, which allowed for recording the Hall voltage with sufficient precision even close to room temperature, where it was fairly small. The good reproducibility of Hall data was proved on another polycrystalline sample (cut from the same ingot) that was notably thicker (0.18 mm), yet had comparable surface dimensions (compare the results in Ref. 12). The electrical leads were fabricated using silver paste to attach 50- μm -thick silver wires to copper pads located on the sample surface. The pads were made by electrochemical deposition of copper from a water solution of copper sulfate. This technique allowed us to reduce the contact resistance down to about 0.1 Ω . The experiments were carried out in the temperature interval 2–300 K and in an applied magnetic field up to 9 T using a PPMS Quantum Design platform. The Hall effect was measured with flipping the position of the specimen in relation to the magnetic field direction using a horizontal rotator. This way the magnetoresistivity component could easily be separated from the Hall signal.

As shown in Fig. 1(a), the Hall coefficient of Ce_2PdIn_8 is negative in the entire temperature and magnetic field ranges studied. In the incoherent Kondo regime, i.e., above $T_{\text{coh}} = 29 \text{ K}$,⁵ R_H is nearly independent of the temperature and the magnetic field strength. Its room-temperature value $R_H^{300\text{K}}$ of $-4.8 \times 10^{-11} \text{ m}^3/\text{C}$ is an order of magnitude smaller than $R_H^{300\text{K}}$ measured for CeCoIn_5 ¹³ and the Ce_2TIn_8 phases with $T = \text{Co, Rh, Ir}$.^{14,15} Within a single-band model, one obtains an unrealistic concentration of electrons being $1.3 \times 10^{29} \text{ m}^{-3}$ (i.e., about $35e^-/\text{f.u.}$). This finding indicates that Ce_2PdIn_8 is a compensated metal, in concert with the results of FS studies of the related compounds.¹⁶⁻¹⁹ Hence the presence of multiple electron and hole bands must be taken into account in reliable calculations of carrier concentrations. In the overall Hall response above approximately 50 K, one may identify a contribution due to skew scattering $R_H^{\text{skew}} = \xi \rho_{\text{mag}} \chi / C$ [see the inset in Fig. 1(a)], where ρ_{mag} is the magnetic contribution to the electrical resistivity, χ is the molar magnetic susceptibility, and C is the Curie constant (the data obtained for the very same polycrystalline specimen of Ce_2PdIn_8 as used in the present study were adopted from Ref. 5). The coefficient ξ represents the magnitude of the

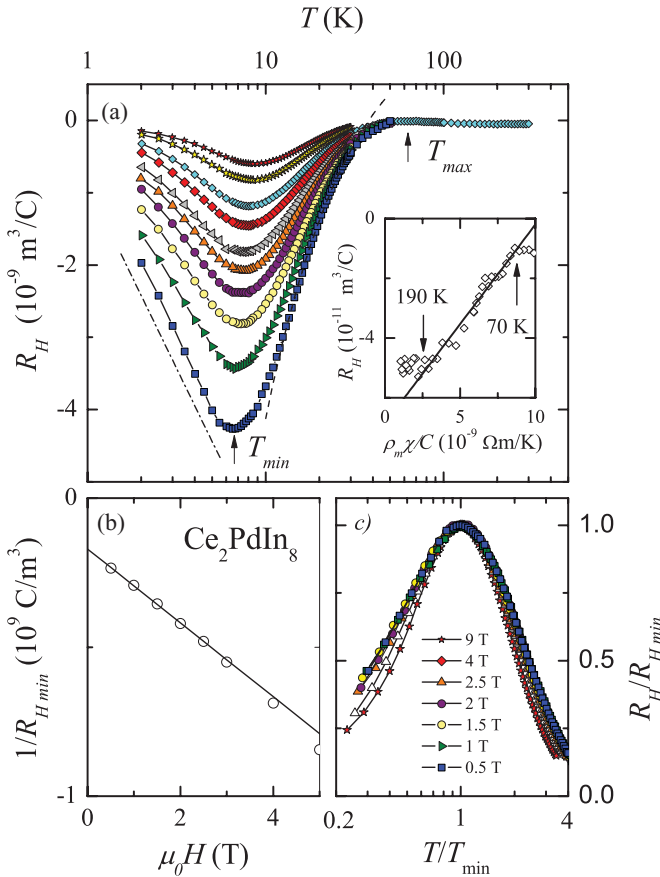


FIG. 1. (Color online) (a) Temperature dependencies of the Hall coefficient of Ce_2PdIn_8 measured in several applied magnetic fields: 0.5, 1, 1.5, 2, 2.5, 4, and 9 T (from the bottom to the top). The dashed and dash-dotted lines illustrate the behaviors discussed in the text. Inset: evaluation of the skew scattering contribution to R_H (see the text). (b) Field variation of $1/R_{H\text{min}}$. The solid line emphasizes the linear dependence observed below 3 T. (c) Normalized Hall data versus normalized temperature (see the text for explanation).

skew scattering effect and may be expressed (for $T \gg T_K$) as $\xi = -\frac{5}{7}g \frac{\mu_B}{k_B} \sin \delta \cos \delta$, where g stands for the magnetic ion Landé factor, μ_B is the Bohr magneton, k_B is the Boltzmann constant, and δ denotes the phase shift produced by incoherent Kondo scattering. The ξ value derived for Ce_2PdIn_8 amounts to only 0.0067 K/T, which implies $\delta = -0.016$ rad. This phase shift is somewhat smaller than those reported for CeCoIn_5 and the other Ce_2TIn_8 phases.^{13–15} Clearly, in contrast to the generic behavior in heavy-fermion systems, the skew scattering mechanism in Ce_2PdIn_8 and the related compounds gives only a minor contribution to the overall Hall response. Although the microscopic origin of this anomalous feature remains unclear, the observed negligible magnitude of R_H^{skew} allows for a detailed analysis of the normal Hall effect.

In the coherent Kondo regime, the Hall coefficient of Ce_2PdIn_8 becomes strongly T and H dependent. In a field of 0.5 T, R_H exhibits a deep minimum at $T_{\text{min}} = 6.5$ K, achieving a very large absolute value of $4.27 \times 10^{-9} \text{ m}^3/\text{C}$ that is nearly two orders of magnitude larger than $R_H^{300\text{K}}$. With increasing the magnetic field strength, the minimum in $R_H(T)$ broadens and slightly shifts to higher temperatures ($T_{\text{min}} \simeq 8.5$ K for $\mu_0 H = 9$ T). Simultaneously, the magnitude

of the Hall response at T_{min} , denoted in the following as $R_{H\text{min}}$, rapidly decreases, especially in weak magnetic fields, and follows the relationship $1/R_{H\text{min}} \propto H$ for fields $\mu_0 H < 3$ T [see Fig. 1(b)]. Interestingly, as can be inferred from Fig. 1(c), plotting the experimental Hall data in the form $R_H/R_{H\text{min}}$ versus T/T_{min} one finds a universal behavior for $\mu_0 H < 3$ T: within the temperature interval $2 \text{ K} \leq T \leq T_{\text{coh}}$, the $R_H(T)$ dependencies collapse onto a single curve. In stronger fields, one observes a gradual deflection of $R_H/R_{H\text{min}}$ from the universal behavior on both sides of T_{min} . Here it is worth noting that T_{min} is very close to the temperature T_{NFL} below which the resistivity of Ce_2PdIn_8 varies linearly with the temperature,⁵ while the Seebeck S/T and Nernst ν/T coefficients,¹² as well as the electronic specific heat coefficient C/T ,^{3,4} exhibit logarithmic divergences. It seems that the observed low-temperature behavior of the Hall response is another indication of the non-Fermi-liquid state in Ce_2PdIn_8 . Possibly the derived scaling of the Hall response represents a generic feature of NFL systems, however this conjecture needs verification from other experimental data and calls for comprehensive theoretical input.

The overall temperature dependence of the Hall coefficient of Ce_2PdIn_8 is strikingly similar to that reported for CeCoIn_5 .¹³ For the latter compound, a theoretical approach has been developed¹ that is based on the microscopic Fermi liquid theory with an important ingredient of the so-called current vertex correction (CVC). It appears that in nearly antiferromagnetic systems there occurs a strong backward scattering of charge carriers due to critical magnetic fluctuations. This mechanism gives rise to an extra current of excited quasiparticles bearing significant momentum dependence, which manifests itself in distinct enhancements in the magnetotransport and thermoelectric effects. In particular, the Hall coefficient exhibits at low temperatures a huge increase in its absolute value, as observed for the CeTIn_5 ($T = \text{Co}, \text{Rh}$) compounds but also for high- T_c cuprates and some organic superconductors.¹ Following the scenario formulated for CeCoIn_5 ,²⁰ the $R_H(T)$ dependence measured for Ce_2PdIn_8 can be divided into three intervals: (i) high-temperature region, where the elastic scattering time is determined by isotropic phonon and incoherent Kondo scattering, and hence R_H is almost T independent, (ii) intermediate-temperature region, for $T_{\text{NFL}} = 8 \text{ K} < T < T_{\text{coh}} = 29 \text{ K}$, where the scattering time is highly anisotropic (because of the presence of hot and cold spots on the Fermi surface) and R_H is dominated by inelastic scattering due to antiferromagnetic fluctuations with a strong CVC effect, and (iii) very low-temperature region, in which the scattering time is governed by isotropic impurity scattering. The electronic transport theory developed by Kontani *et al.*¹ assumed a 2D character of the Fermi surface, and hence it predicted for the (ii) interval a $R_H \propto \xi_{\text{AF}}^2 \propto T^{-1}$ dependence (ξ_{AF} is the AF correlation length), which has indeed been experimentally observed for CeCoIn_5 .²⁰ In the case of Ce_2PdIn_8 , this type of functional dependence of R_H appears less obvious. The relation predicted for a 2D AF system can be found in a rather narrow temperature range $12 \text{ K} < T < 30 \text{ K}$ [see the dashed line in Fig. 1(a)], yet only if a more general formula $R_H = R_H^0 + A/(\theta + T)$ is applied. On the other hand, a similarly good description of the Hall data in this temperature range can be obtained using a function $R_H \propto A/(\theta + T)^{3/2}$, which has been found within

the self-consistent renormalization theory for a purely 3D AF system.²⁰ Here it is worth recalling that Ce_2TIn_8 phases are believed to have less distinct 2D character than their CeTIn_5 counterparts, as their crystallographic unit cells consist of two 3D CeIn_3 units and a single 2D slab of TIn_2 .²¹

According to Ref. 20, in CeCoIn_5 and related phases the region of predominance of the anisotropic scattering ceases at the temperature below which $R_H(T)$ becomes mainly governed by the isotropic impurity scattering. However, for NFL systems one should generally also consider a different scenario that accounts for temperature-dependent changes in the quasiparticle effective masses m^* . The logarithmic divergence of the electronic contribution to the specific heat, observed for Ce_2PdIn_8 below $T_{\text{NFL}} \simeq 8$ K,^{5,9} implies $m^* \sim C/T \sim \ln T$. Accordingly, one might expect that in a similar temperature region, the Hall coefficient $R_H \sim 1/m^*$ varies inversely proportional to $\ln T$. Interestingly, it seems that such a temperature dependence of R_H indeed occurs below $T_{\text{min}} \simeq T_{\text{NFL}}$ in the weak magnetic field limit [see dash-dotted line in Fig. 1(a)]. Recently, an approach called fermion condensation quantum phase transitions theory (FCQPT) has been derived for NFL systems, which explicitly takes into account m^* dependencies on temperature, magnetic field, and/or charge carrier density.²² It seems tempting to numerically apply the FCQPT model to the magnetotransport data of Ce_2PdIn_8 .

Figure 2(a) presents the magnetic field variations of the transverse magnetoresistivity [defined as $MR(H) = [\rho_{xx}(H) - \rho_{xx}(0)]/\rho_{xx}(0)$] measured for Ce_2PdIn_8 in the temperature interval 2–30 K, i.e., in the coherent Kondo regime. In the entire temperature range studied, MR is positive, and its magnitude increases with increasing field strength, reaching about 18.2% at 4 K in a field of 9 T. The observed convex curvature of the magnetoresistivity isotherms, which is more evident the lower the temperature, suggests the development of a negative contribution in strong magnetic fields. This effect likely arises from gradual suppression of spin-flip scattering by the magnetic field. One might expect that a broad maximum in $MR(H)$ would develop at temperatures lower than 2 K, with its position being shifted to smaller fields with rising temperature, as observed, e.g., for CeCoIn_5 .²⁰ Interestingly, as shown in Fig. 2(b), the experimental MR data can be plotted as the normalized magnetoresistivity $MR(H)/MR(H^*)$ versus the normalized field H/H^* to get a universal curve over an extended interval in the magnetic field strength. The scaling parameter H^* , which provides an excellent overlap of all the isotherms, appears independent of the temperature above $T_{\text{min}} \simeq T_{\text{NFL}} = 8$ K, and decreases linearly with T at lower temperatures [see the inset to Fig. 2(b)], i.e., in the range where the electrical resistivity,⁵ the specific heat,^{3,4} the thermoelectric coefficients,¹² and the Hall coefficient of Ce_2PdIn_8 exhibit distinct non-Fermi-liquid character. This coincidence suggests that the revealed behavior of $H^*(T)$ is another indication of the onset of quantum criticality in the compound studied. Hypothetically, such behavior of the magnetoresistivity might be universal for systems at the verge of magnetic instability.

In the presence of strong backflow scattering of charge carriers on critical AF spin fluctuations, one can expect a violation of the Kohler's rule $MR = F(H/\rho_{xx})$ that is known to be well applicable to most FL systems.²³ Instead, for CeCoIn_5 and other CeTIn_5 compounds with a significant CVC

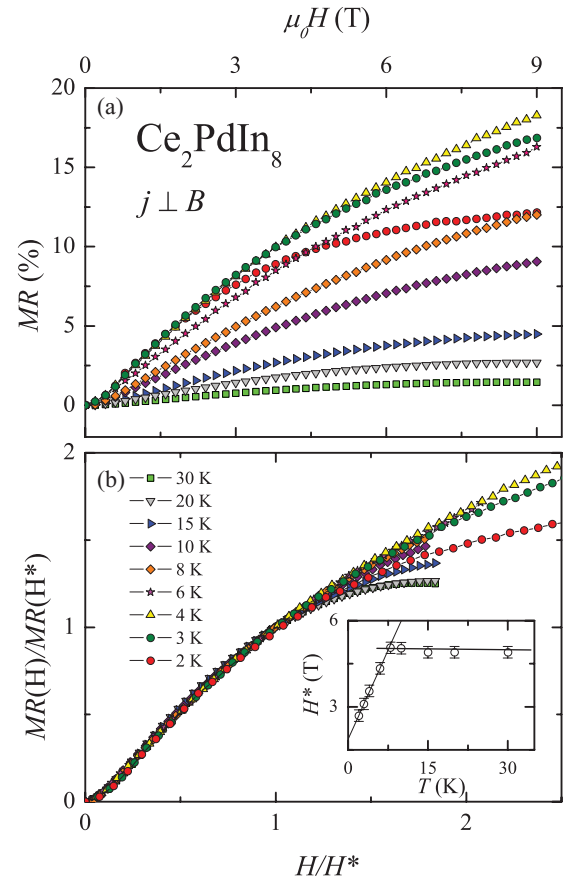


FIG. 2. (Color online) (a) Transverse magnetoresistivity isotherms of Ce_2PdIn_8 taken at a few different temperatures above 2 K. (b) Normalized magnetoresistivity data plotted versus normalized field (see the text). Inset: temperature dependence of the scaling parameter H^* .

contribution, a different scaling has been validated,²⁰ namely $MR \propto \tan^2 \Theta_H$, where $\Theta_H = \tan^{-1}(\rho_{xy}/\rho_{xx})$ is the Hall angle. Remarkably, the latter so-called modified Kohler's rule (MKR) has also been experimentally observed for several high- T_c superconductors.²³ The results [displayed in Fig. 3(a)], clearly show that the Kohler's scaling does not work for Ce_2PdIn_8 , however in some extended ranges of the parameters, the MR data is proportional to $\tan^2 \Theta_H$ [see Fig. 3(b)]. Moreover, in weak magnetic fields, the MR isotherms measured in the NFL region (i.e., below 8 K) converge into a single curve that may approve the MKR approach for this compound.

In summary, in the coherent state, i.e., for $T_{\text{coh}} < 29$ K, the Hall coefficient of Ce_2PdIn_8 strongly depends on both the temperature and the magnetic field strength, while the transverse magnetoresistivity seems to obey the modified Kohler's rule, at least in the weak field limit. These anomalous magnetotransport properties exhibit close similarity to those reported before for CeCoIn_5 ,²⁰ and can be explained in terms of the recently developed theory in which anisotropy of the Fermi surface and strong backflow effect are taken into account.²³ Undoubtedly, the low-temperature normal-state electrical behavior of Ce_2PdIn_8 is governed by AF fluctuations, which eventually give rise to the onset of the unconventional superconductivity emerging near AF QCP.^{6,8,9} The derived

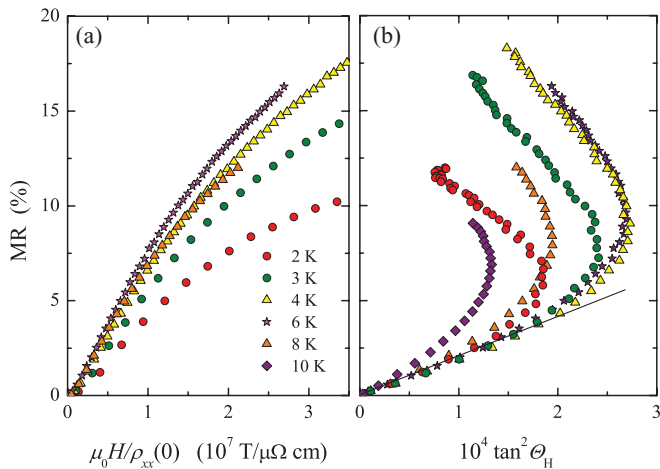


FIG. 3. (Color online) Scaling of the transverse magnetoresistivity of Ce_2PdIn_8 according to (a) Kohler's rule and (b) the modified Kohler's rule (see the text for explanation). The straight line in (b) emphasizes a universal behavior.

scalings of the Hall effect data, $R_H(T)/R_H$ versus T_{\min} and $R_{H\min}$ versus H , suggest the occurrence in the NFL state of a characteristic magnetic scale of the order of the upper critical field $H_{c2}(0)$. Furthermore, in the extended temperature and magnetic field ranges, the normalized transverse magnetoresistivity of Ce_2PdIn_8 has been found to exhibit a relationship $\text{MR}(H)/\text{MR}(H^*)$ versus H/H^* with a sole scaling parameter H^* that is proportional to the temperature in the NFL state (below $T_{\text{NFL}} = 8$ K) and T independent at higher temperatures (at least up to $T_{\text{coh}} = 29$ K). It remains to be verified, both experimentally and theoretically, if the magnetotransport scalings observed for Ce_2PdIn_8 have universal applicability to heavy-fermion NFL systems close to AF QCP instability.

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