Superconductivity and non-Fermi-liquid behavior of Ce_2PdIn_8

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The electrical resistivity of the heavy fermion superconductor Ce_2PdIn_8 was measured in magnetic fields up to 12 T and under hydrostatic pressures up to 21 kbar. At zero field, the low-temperature electrical resistivity in the normal state exhibits a power-law behavior $\rho_0 + AT^n$ with $n < 2$. In this non-Fermi-liquid regime, both the superconducting temperature T_c and the coefficient A decrease with increasing pressure, while the exponent n and the resistivity maximum at T_{max} increase. The findings indicate a destabilization of the superconducting state via increasing hybridization strength between the 4*f* and conduction electrons. In concert, enlargement of the *f*-band width at the Fermi level results in a decrease of the density of states $N(E_F)$. Application of magnetic fields recovers the Fermi-liquid state at H_{c2}, at which both A and ρ_0 show a tendency to diverge. The data obtained indicate that any change in the Kondo interaction strength in Ce_2PdIn_8 by applied pressure or quenching spin fluctuations by external magnetic fields results in pushing away the system from the non-Fermi-liquid regime concomitantly with the destruction of the superconducting state. These new results support a scenario in which the superconductivity in Ce_2PdIn_8 is driven by antiferromagnetic spin fluctuations in the vicinity of an underlying quantum critical point.

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I. INTRODUCTION

In metallic cerium compounds, the competition between the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and the Kondo effect is usually assumed to be responsible for a variety of physical phenomena. This concept was discussed a long time ago by Doniach,[1] in terms of the function $|JN(E_F)|$, where $|J|$ is the magnitude of the magnetic exchange interaction and $N(E_F)$ is the electronic density of states at the Fermi energy E_F. When the RKKY interaction dominates over the Kondo effect, real systems show magnetic order. In the opposite situation, nonmagnetic ground states are found. Between these two extreme situations, there exists a critical region, where exotic phenomena such as a heavy fermion (HF) behavior, non-Fermi-liquid (nFL) features, or a quantum critical point (QCP) are expected to occur.[2–4] Several experiments have proved that on approaching the critical region from the magnetically ordered side, superconductivity may appear.[5–8] This type of superconductivity is not very well understood because it coexists with weak magnetism or situates near QCPs, suggesting new pairing mechanisms, different from the BCS electron-phonon attraction.

Recently, the intermetallic compound Ce_2PdIn_8 was found to exhibit a superconducting transition at $T_c = 0.7$ K.[9–13] Measurements of the bulk properties of polycrystalline and single-crystalline samples yielded the upper critical field $H_{c2}(0) \approx$ 2.5 and 4.8 T, respectively. Similarly, the Ginzburg-Landau coherence length was estimated to be $\xi \approx 8.2$ and 11.5 nm, respectively, and the magnetic penetration depth $\lambda \approx 170$ and 400 nm, respectively. A large ratio $l/\xi_0 \approx 8$–40 between the mean free path and the BCS coherence length indicates that Ce_2PdIn_8 is a clean limit superconductor. Moreover, it has been shown that unconventional superconductivity in this material is closely associated with the HF properties. The Sommerfeld coefficient measured at 0.3 K in a magnetic field of 5 T [i.e., larger than $H_{c2}(0)$] is as large as 500 mJ/K^2 mol Ce, which yields an effective mass of $\sim 200m_0$. Interestingly, just above the onset of the superconducting state the physical properties of Ce_2PdIn_8 are typical for nFL systems. The compound shows a linear temperature dependence of the electrical resistivity $\rho(T) \propto T$ and a logarithmic dependence of the electronic specific heat $C/T(T) \propto \ln T$ in a wide temperature range.[9,12] All these hitherto reported data suggested that Ce_2PdIn_8 is very similar to the structurally closely related compounds $CeCoIn_5$, $CeIrIn_5$, and Ce_2CoIn_8,[14–16] in which superconductivity is believed to situate near a QCP. In order to clarify the relationship between unconventional superconductivity and the nFL state associated with a QCP, it is necessary to investigate the superconducting state and the normal state in Ce_2PdIn_8 in more detail.

In this paper we report on the effect of applying hydrostatic pressure, P, and magnetic fields, H, on the electrical resistivity of a high-quality polycrystalline Ce_2PdIn_8 specimen. Both these external P and H parameters were found to result in a decrease of T_c and in driving the system away from the nFL state. We argue that pressure increases J owing to an enhancement in the hybridization strength between conduction electrons and f electrons, and simultaneously it decreases $N(E_F)$ owing to an enlargement of the *f*-band width at the Fermi level. On the other hand, the application of magnetic fields $H < H_{c2}$ (for a given value of pressure) yields an enhancement of spin fluctuations and thus leads to an increase of the quasiparticle effective mass. In stronger fields (above H_{c2}), spin fluctuations become suppressed and the low-temperature properties of the compound are those of a Fermi liquid. Based on the obtained results we conclude that superconductivity in Ce_2PdIn_8 is mediated by antiferromagnetic (AF) spin fluctuations associated with an underlying QCP.

II. EXPERIMENTAL

A polycrystalline sample of Ce_2PdIn_8 was prepared by arc melting the stoichiometric amounts of high-purity elemental components (Ce-3N, Ames Laboratory, Pd-3N, Chempur and

In-6N, Chempur). Details on the synthesis were given in Ref. 12. The quality of the product was proved by powder x-ray diffraction and microprobe analysis. The derived lattice parameters were nearly same as those reported previously.[12] No foreign phases were found, in particular, $CeIn_3$ or metallic In. Moreover, the high quality of the studied sample was approved by a rather large value of the residual resistivity ratio RRR = 12, notably larger that those reported for single crystals.[9,13]

The electrical resistivity under pressure up to 21 kbar was measured in the temperature range 0.3–250 K. The measurements were performed with a four-point ac bridge technique and a current of 1 mA. Hydrostatic pressure was generated by a piston cylinder cell using Daphne oil as a pressure-transmitting medium. In all the measurements, pressure was fixed at room temperature and the data were taken on cooling the specimen with a rate of 0.5 K/min. High-quality Pb was used as a pressure indicator; its superconducting transition width was less than 30 mK. At selected pressures, transverse magnetoresistance measurements were performed in magnetic fields up to 12 T using a ^{3}He cryostat equipped with a Cryogenics LTD magnet.

III. RESULTS AND DISCUSSION

The temperature dependencies of the electrical resistivity of Ce_2PdIn_8 as measured under various pressures are shown in Fig. 1. Upon application of pressure, the room temperature resistivity (RTR) initially jumps to a higher value and then its magnitude varies linearly with a rate 0.37 $\mu\Omega$ cm/kbar. The behavior observed at 4.5 kbar is likely mainly owing to electrical contact sliding inside the pressure cell, yet some contribution owing to grain boundaries and other crystal defects is also possible. Remarkably, a very similar pressure response was observed for $CeCoIn_5$, where after an initial jump, the RTR also follows a linear P dependence with a rate of $\approx$0.4 $\mu\Omega$ cm/kbar.[17] The linear slope of RTR(P) revealed for the two indides is rather small [it may be compared, e.g., with those reported for $CeCu_2Si_2$ (2.8 $\mu\Omega$ cm/kbar) (Ref. 18) and CePtSi (1.6 $\mu\Omega$ cm/kbar) (Ref. 19)], which indicates that

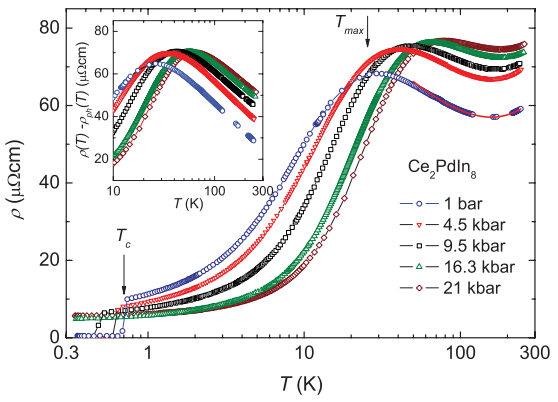

FIG. 1. (Color online) Electrical resistivity of Ce_2PdIn_8 as a function of temperature under several hydrostatic pressures up to 21 kbar. The solid line is a fit of the data to Eq. (1). The inset represents the resistivity at approximately T_{max}.

in both compounds the phonon contribution to the resistivity is hardly affected by pressure.

An inspection of the $\rho(T,P)$ curves shows that the data taken at 1 bar are very similar to those previously reported for ambient pressure.[12] It exhibits a maximum at T_{max}, which is a feature characteristic of an interplay between Kondo and crystalline electric-field interactions. Above T_{max}, $\rho(T)$ varies as $-\ln T$ owing to incoherent Kondo scattering of the conduction electrons, while below T_{max} a rapid decrease of the resistivity with decreasing temperature occurs owing to the onset of coherent scattering of the conduction electrons from Kondo centers arranged periodically in the crystal lattice. In order to determine precisely the values of T_{max} at different pressures, the phonon contribution should be subtracted from the measured resistivity. Assuming that the high-temperature phonon scattering is given by $\rho_{ph} = c_{ph}T$ and the incoherent Kondo scattering follows the function $\rho_K = -c_K \ln T$, the high-temperature resistivity data can be described by

$$\rho(T) = \rho_0 + \rho_\infty + \rho_{ph} + \rho_K, \qquad (1)$$

where ρ_0 and ρ_∞ are the resistivity terms owing to scattering the conduction electrons on static defects in the crystal lattice and on disordered magnetic moments, respectively. As an example, a least-squares fit of Eq. (1) to the experimental data taken at 1 bar is depicted in Fig. 1 by the solid line. A similarly good description of the high-temperature resistivity in terms of Eq. (1) was obtained for the other pressures, hence supporting the presence of the Kondo interaction in the compound studied. Shown in the inset to Fig. 1 is the nonphononic contribution to the resistivity $\rho(T)$-ρ_{ph}. Apparently, with increasing pressure the T_{max} value systematically shifts to higher temperatures. In the pressure range studied, T_{max} is $\sim$25 K at 1 bar and increases linearly with a rate $dT_{max}/dP = 1.8$ K/kbar, reaching $\sim$62 K at 21 kbar [see Fig. 2(a)]. This behavior is similar to that reported for the $CeTIn_5$ superconductors, which also exhibit a maximum in $\rho(T)$.[16,17,20,21] For example, in $CeCoIn_5$ $T_{max} \approx 50$ K (ambient pressure) changes with a rate of $\sim$2.8 K/kbar.[17] In turn, in $CeRhIn_5$, $T_{max} \approx 23.5$ K was observed at 13.5 kbar, and for higher pressures this maximum shifts to higher temperatures with the rate $dT_{max}/dP = 1$ K/kbar.[20] For all these compounds, T_{max} corresponds to the coherence temperature T_{coh}.[22] An increase of T_{coh} upon applying pressure is naturally expected in cerium compounds because pressure leads to an enhancement of the hybridization between the conduction electrons and the f electrons (represented by J), and simultaneously results in an enlargement of the f-band width. The latter causes a reduction of the density of states at the Fermi level $N(E_F)$. Thus, it seems fully justified presuming that a very similar pressure response occurs in the closely related indide Ce_2PdIn_8.

Application of hydrostatic pressure remarkably influences the position of T_{max} but causes a rather weak change in the Kondo term represented by the resistivity slope $c_K = -d\rho(T)/d\ln T$ [see Fig. 2(b)]. Within the model developed by Cornut and Coqblin,[23] the parameter c_K characterizes the Kondo interaction strength and depends on $|J^3N(E_F)^2|$. The decrease of the parameter c_K indicates that $|J^3N(E_F)^2|$ decreases with pressure. Considering the pressure dependence of T_{max} and c_K, one may conclude that $N(E_F)$ strongly diminishes with increasing pressure.

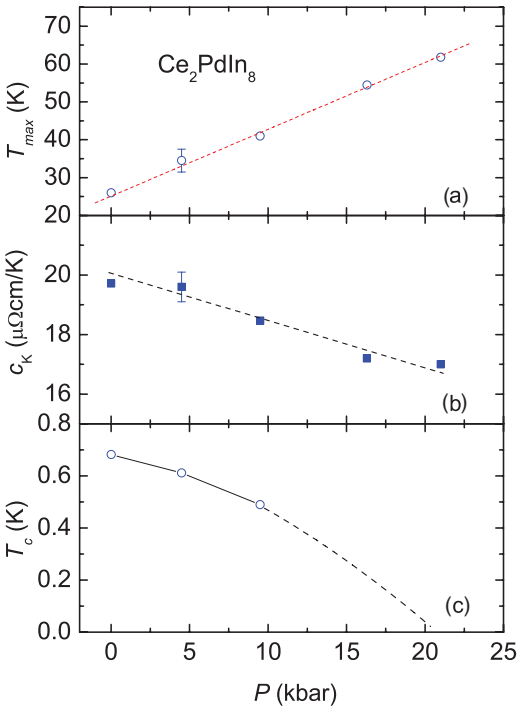

FIG. 2. (Color online) Pressure dependencies of (a) the position of the maximum in the electrical resistivity, (b) the Kondo coefficient $c_K = -d\rho(T)/d\ln T$, and (c) the critical temperature T_c.

The pressure dependence of the superconducting transition in Ce_2PdIn_8 is shown in Fig. 2(c). At 1 bar T_c is equal to 0.7 K, in agreement with the previous report,[9,12] and decreases with increasing pressure. For pressures higher than 16 kbar, the onset of superconductivity is not observed anymore above 0.35 K. A rough extrapolation of the T_c data suggests that superconductivity vanishes at $P \sim 21$ kbar. It is worthwhile noting that the pressure dependence of T_c in Ce_2PdIn_8 remarkably differs from those of $CeCoIn_5$ and $CeRhIn_5$. For the latter compounds, a superconducting dome in the T-P phase diagram was observed, in which T_c achieves its maximum value at a critical pressure P^* of ~13 kbar (Ref. 24) and 23 kbar,[25] respectively. It was also shown that P^* separates two distinct regimes, namely, (i) nFL characteristics up to P^* and (ii) Fermi-liquid-like properties above P^*.[24] In the case of Ce_2PdIn_8, as discussed below, the Fermi-liquid state has not been observed in the pressure range studied, and superconductivity develops only from the nFL state.

Figure 3 displays the low-temperature electrical resistivity of Ce_2PdIn_8. Remarkably, the resistivity measured just above T_c shows no T^2-dependence characteristic of the Fermi-liquid state. Instead, $\rho(T)$ exhibits a power-law T^n dependence with $n < 2$, which is usually considered as a fingerprint of a nFL state. A possible origin of this behavior is scattering of conduction electrons on two-dimensional antiferromagnetic quantum fluctuations.[26] Results of least-squares fits of $\rho(T) = \rho_0 + AT^n$ in the temperature range $T_c - 5$ K are shown in Fig. 3 as solid lines. Values of the residual resistivity ρ_0, the exponent n, and the coefficient A are presented in Fig. 4. For the pressure range studied, n was found to increase from 1 at ambient pressure to 1.3 at 21 kbar. Previously, similar power-law exponents were found in magnetic heavy-

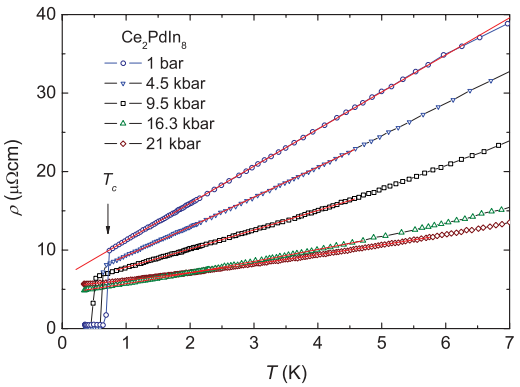

FIG. 3. (Color online) The electrical resistivity under pressures near the superconducting transition. The solid lines represent the fits of the equation $\rho(T) = \rho_0 + AT^n$.

fermion systems, tuned by pressure to superconductivity,[5] in $CeTIn_5$ compounds[27] and in high-T_c cuprates.[28] Remarkably, both n and ρ_0 start to increase at $P \sim 21$ kbar, coinciding with the suppression of superconductivity near this value of pressure [cf. Fig. 2(c)]. This hints to a superconducting state in Ce_2PdIn_8 being directly related to a quantum critical behavior.

As is apparent from Fig. 4, the coefficient A rapidly decreases with increasing pressure, from 4.7 $\mu\Omega$ cm/K at 1 bar down to 0.6 $\mu\Omega$ cm/K$^{1.3}$ at 21 kbar. In a number of heavy-fermion compounds, the prefactor A in $\rho(T) = \rho_0 + AT^2$ is

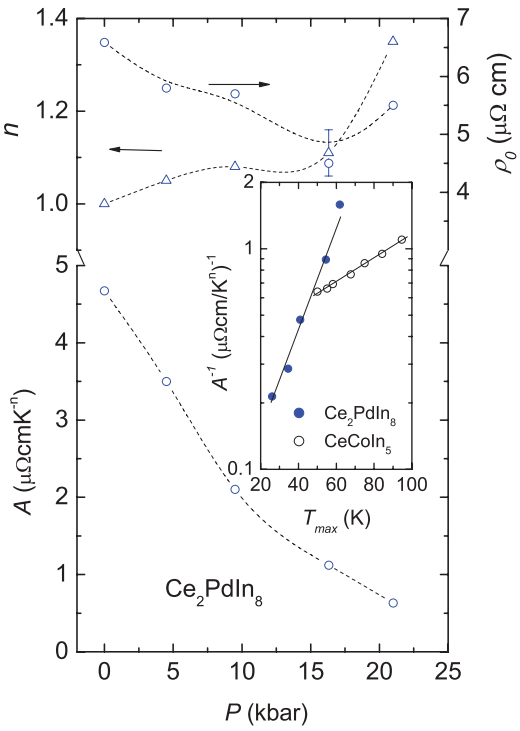

FIG. 4. (Color online) Pressure dependencies of the exponent n (left-hand scale), the residual resistivity ρ_0 (right-hand scale), and the resistivity prefactor A (bottom). The dashed lines serve as a guide to the eye. The inset shows the relationship between A^{-1} and T_{max}. The data of $CeCoIn_5$ are taken from Refs. 17 and 24. The solid lines emphasize a linear dependence of $\ln(A^{-1})$ vs T_{max}.

proportional to the square of the electronic specific heat γ ($A \propto \gamma^2$) and it is also proportional to the inverse of the square of Kondo temperature ($A \propto T_K^{-2}$).[29] For nFL systems, no similar relationships between A, γ, and T_K have been worked out yet. Nevertheless, the experimental data derived for $CeCoIn_5$ seem to indicate a proportionality between A and γ derived in the nFL regime.[24] A similar relationship likely holds for Ce_2PdIn_8 for which a clear reduction of $N(E_F)$ with increasing pressure was observed (see above). In the inset to Fig. 4, the values of A^{-1} are plotted as a function of T_{max} determined from the high-temperature resistivity data. Clearly, a proportionality $\ln(A^{-1}) \sim T_{max}$ is observed for the entire pressure range studied. Moreover, a similar relationship is observed also for $CeCoIn_5$. It seems quite remarkable that the coefficient A in both nFL compounds correlates with T_{max}, which can be associated with the coherence temperature T_{coh} (see above), rather than showing any straight correlation with T_K that is a single-ion Kondo temperature in Fermi-liquid systems. This unique type of scaling should be tested for other nFL systems.

For several hydrostatic pressures, the electrical resistivity of Ce_2PdIn_8 was measured in magnetic fields up to 12 T, and applied perpendicular to the electrical current (transverse magnetoresistance). As an example, Fig. 5 presents the data collected at 1 bar; similar results were obtained at higher pressures. The resistivity maximum at T_{max} was found to be hardly field dependent for fields $\mu_0 H < 3$ T. However, above 3 T the maximum in $\rho(T)$ shifts toward lower temperatures with increasing field strength [see Fig. 5(a)]. This behavior can be rationalized as suppression of spin fluctuations by the magnetic field, which manifests itself in a gradual reduction

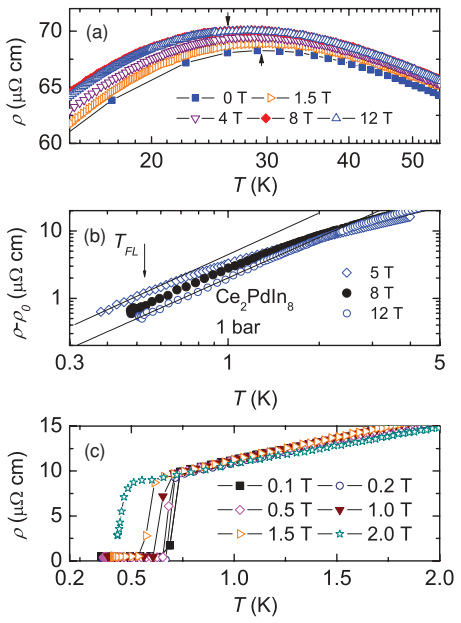

FIG. 5. (Color online) Temperature variations of the electrical resistivity of Ce_2PdIn_8 measured at ambient pressure in various applied magnetic fields. (a) The data around the resistivity maximum at T_{max} (denoted by the arrows). (b) Low-temperature data taken in $H > H_{c2}$. The solid lines indicate a AT^2 dependence below the Fermi-liquid temperature T_{FL} (denoted by the arrow). (c) Low-temperature data taken in $H < H_{c2}$.

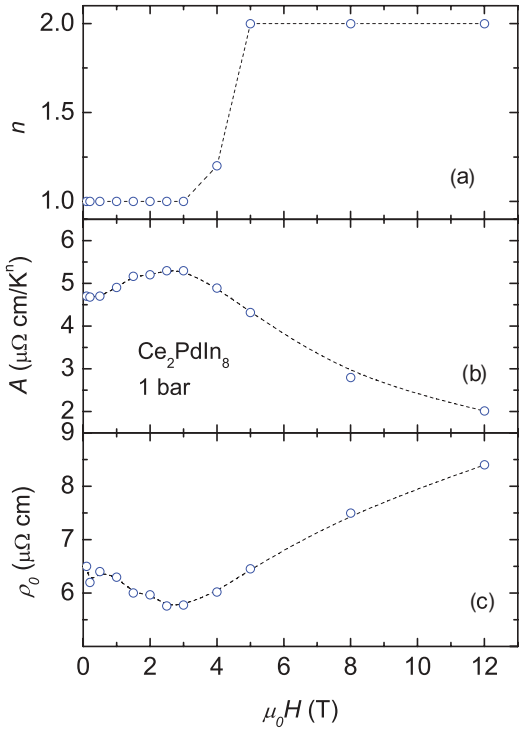

FIG. 6. (Color online) Field dependencies of the exponent n, the coefficient A, and the residual resistivity ρ_0 obtained from the fits of the equation $\rho(T) = \rho_0 + AT^n$ to the low-temperature resistivity data.

of the coherence temperature $T_{coh} \approx T_{max}$. When comparing the normal-state resistivity of Ce_2PdIn_8 measured in fields above [Fig. 5(b)] and below [Fig. 5(c)] $\mu_0 H_{c2} \approx 2.5$ T, one recognizes a drastic change in the slope $d\rho(T)/dT$ when the applied field is tuned through H_{c2}. Below H_{c2}, the normal-state resistivity can only be represented by the aforementioned nFL function $\rho(T) = \rho_0 + AT$. In contrast, $\rho(T)$ measured in fields above H_{c2} can be described by the power law $\rho \sim AT^2$ in the temperature range $0.3 - T_{FL}$, where T_{FL} increases with rising field strength. The observation of a T^2 dependence of the resistivity indicates that the Fermi-liquid ground state in Ce_2PdIn_8 is recovered by application of strong enough magnetic fields. Figure 6 summarizes the field-dependent values of n, A, and ρ_0. Apparently, all these parameters exhibit anomalies near H_{c2}, namely, n jumps to $n = 2$, A shows a maximum, while $\rho_0(H)$ goes through a minimum. All these findings corroborate the scenario of AF spin fluctuations associated with a QCP, which govern the behavior of Ce_2PdIn_8 close to H_{c2}. Below the critical field, spin fluctuations become enhanced on approaching the AF QCP, whereas above H_{c2} they are gradually suppressed by field.

The magnetic-field and hydrostatic pressure dependencies of T_c and T_{FL} allow constructing a P-H-T phase diagram shown in Fig. 7. At 1 bar, the upper critical field $H_{c2}(0)$, taken as the value extrapolated to zero temperature, is ~2.5 T. The initial slope of the upper critical field at T_c, $-\mu_0 dH_{c2}/dT$, amounts to 16(1) T/K. The coherence length can be estimated as $\xi \approx 10.5$ nm by using the relation $H_{c2} = \Phi_0/2\pi\xi^2$, where Φ_0 is a quantum flux. Similarly, from the data measured at 4.5 kbar, one obtains the characteristic superconducting

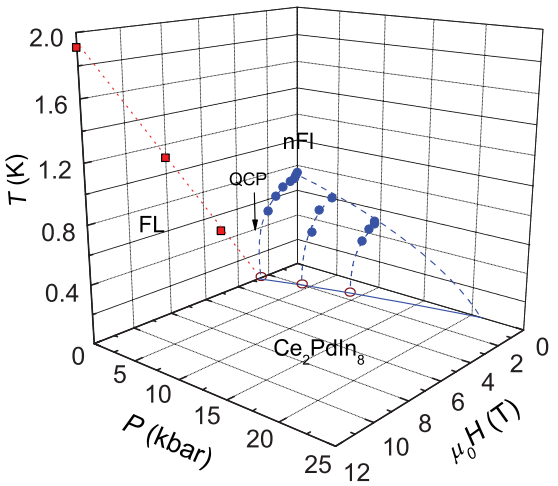

FIG. 7. (Color online) *T-P-H* phase diagram of a polycrystalline Ce_2PdIn_8. The closed circles denote T_c, the squares represent T_{FL}, and the open circles stand for H_{c2}. The dashed and dotted lines serve as a guide for the eye. The solid curve is a hypothetical QCP line.

parameters, $\mu_0 H_{c2} = 2.2$ T, $-d\mu_0 H_{c2}/dT = 15(1)$ T/K, and $\xi \approx 12.2$ nm, while those derived from the the 9(1) kbar data are $\mu_0 H_{c2} = 1.8$ T, $-d\mu_0 H_{c2}/dT = 9.5$ T/K and $\xi \approx 13.5$ nm. In terms of clean superconductors,[30,31] the initial slope is related to the effective mass of the superconducting carriers as $-dH_{c2}/dT \propto (m^*)^2 T_c$. Hence, the observed change in the values of $-dH_{c2}/dT$ with increasing pressure indicates that m^* is likely at its maximum at pressures below 4.5 kbar.

Finally, we should emphasize that there exists a distinct difference in the values of $\mu_0 H_{c2}$ obtained for single-crystalline[9] and polycrystalline samples of Ce_2PdIn_8. This is an indication of strongly anisotropic properties of the investigated compound. For that reason the absolute values of the transport coefficients (e.g., A, c_K, ρ_0) derived from the data obtained on polycrystalline material must be taken with appropriate caution. However, the pressure and magnetic-field dependencies of these quantities, demonstrated in this work, are believed to represent the actual behavior of the compound studied.

IV. SUMMARY

The temperature-dependent electrical resistivity of a good-quality polycrystalline sample Ce_2PdIn_8 obtained in magnetic fields up to 12 T and under hydrostatic pressures up to 21 kbar indicate that heavy-fermion superconductivity in this compound emerges from a nFL normal state. Application of pressure and/or sufficiently strong magnetic fields results in a suppression of superconductivity and simultaneously pushes the system away from the nFL state. In the pressure range available there is no evidence of a Fermi-liquid behavior nor of any long-range magnetic ordering. A Fermi-liquid state, however, is recovered in magnetic fields above the critical field $\mu_0 H_{c2}$, which seems to be a quantum critical point. The pressure and field dependencies of the resistivity suggest that superconductivity in Ce_2PdIn_8 is likely mediated by the AF spin fluctuations in the very proximity of an avoided quantum critical phase transition.

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