Quantum criticality near the upper critical field of Ce$_2$PdIn$_8$

Y. Tokiwa and P. Gegenwart

I. Physikalisches Institut, Georg-August-Universität Göttingen, 37077 Göttingen, Germany

D. Guida and D. Kaczorowski

Institute of Low Temperature and Structure Research, Polish Academy of Sciences, PO Box 1410, 50-950 Wroclaw, Poland

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We report low-temperature specific heat measurements in magnetic fields up to 12 T applied parallel and perpendicular to the tetragonal c-axis of the heavy fermion superconductor Ce$_2$PdIn$_8$. In contrast to its quasi-two-dimensional (2D) relative CeCoIn$_5$, the system displays an almost isotropic upper critical field. While there is no indication for a FFLO phase in Ce$_2$PdIn$_8$, the data suggest a smeared weak first-order superconducting transition close to $H_{c2}$ $\approx$ 2 T. The normal state electronic specific heat coefficient displays logarithmically divergent behavior, comparable to CeCoIn$_5$ and in agreement with 2D quantum criticality of spin-density-wave type.

Unconventional superconductivity (SC) often occurs in systems with competing phases near a quantum critical point (QCP). Indicative for quantum criticality is e.g. the observation of non-Fermi liquid (NFL) behavior in the normal state at $T > T_c$ in zero field or down to the lowest accessible temperatures at fields above the upper critical magnetic field $H_{c2}$. Heavy-fermion (HF) superconductors have sufficiently small $H_{c2}$, allowing for a detailed thermodynamic study of the normal state properties down to very low temperatures to investigate NFL behavior. The layered tetragonal CeCoIn$_5$ with $T_c = 2$ K is a prototype heavy-fermion superconductor with a NFL normal state due to a field-induced QCP slightly below $H_{c2}$. Its SC properties are also fascinating. The transition at $H_{c2}(T)$ becomes first order below 1 K due to strong Pauli limiting and clear phase transition anomalies at the high-field and low-temperature (HFLT) corner in the SC phase diagram have been discovered which were thought to be related to a modulated SC Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state. The origin of the HFLT SC state is under extensive debate, as subsequent NMR and neutron diffraction experiments have revealed the existence of an antiferromagnetic (AF) ordering within the HFLT phase while a very recent NMR study suggests spatially distributed normal quasi-particle regions due to the formation of a FFLO state.

The recently discovered HF superconductor ($T_c = 0.7$ K) Ce$_2$PdIn$_8$ (Ref. 19) belongs to the same class of Ce$_n$TIn$_{3n+2}$ (T; transition metal, n = 1, 2 and $\infty$) systems containing a series of CeIn$_3$ and Tin layers stacked along the c-axis. While the $n = 1$ systems with $T$=Co, $T$=Ir (Ref. 22) and $T$=Rh (Ref. 22) display anisotropic properties due to the layered, quasi two-dimensional (2D) structure, cubic CeIn$_3$ (n = $\infty$) is a completely isotropic system. The bilayer ($n = 2$) systems are therefore expected to display intermediate behavior due to the more 3D character of their crystal structure consisting of a stack of two CeIn$_3$ layers separated by one PdIn$_2$ layer. Ce$_2$PdIn$_8$ is an ambient-pressure HF superconductor of this class of materials. Its upper critical field $H_{c2}$ is strongly Pauli limited with a large Maki parameter of 2.9. Clean single crystals have become available, with a residual resistivity of about 2.5 $\mu$$\Omega$cm and a large mean free path of $l=420 \AA$ exceeding the SC coherence length $\xi=82 \AA$. Thus, the system fulfills the necessary conditions for the formation of the FFLO state and it is interesting to investigate the high-field and low-temperature part of the SC phase by a thermodynamic probe. Furthermore, recent low-T electrical resistivity measurements at fields above the upper critical field have revealed strong similarities to CeCoIn$_5$, i.e. a quasi-linear dependence at $H_{c2}$ which turns into Fermi liquid behavior only at larger fields suggesting a field-induced QCP near $H_{c2}$. Because of these common features, it is highly interesting to study the low-temperature specific heat of single crystalline Ce$_2$PdIn$_8$ in magnetic fields applied both parallel and perpendicular to the tetragonal c-axis.

Single crystals were grown by the self-flux method and very thin high-quality pieces were glued on the sample holder. The basal plane orientation for the different pieces was not investigated. The specific heat at temperatures down to 70 mK and magnetic fields up to 12 T was measured in a dilution refrigerator with a SC magnet by employing the quasiadiabatic heat pulse method.

Figure 1 shows the specific heat after subtraction of the nuclear contribution as $C_{el}/T$, at magnetic fields up to $H_{c2}$. The nuclear contribution has been determined from the $\alpha/T^3$ contribution to $C/T$ at lowest temperatures. We have verified, that $\alpha \sim H^2$ as expected for paramagnetic materials. At zero field, $C_{el}/T$ increases upon cooling to $T_c$ = 0.68 K, at which a typical mean-field type anomaly is found. Within the SC state, it decreases linearly down to 0.2 K and saturates at a residual value of $C_{el}/T = 0.65$ J/mol K$^2$ (0.33 J/Ce-mol K$^2$), which is much larger than the respective value of 0.04 J/mol K$^2$ found for CeCoIn$_5$, indicating a larger quasi-particle density of states arising from pair breaking of defects. The specific heat at magnetic fields along different directions is rather similar and displays only a weak anisotropy in $H_{c2} = 2.0$ T and $2.2$ T for $H \perp |001|$ and $H |||001|$, re-
spectively. At high fields, the SC transition changes its shape and becomes a rather symmetric peak different to the expectation for standard superconductors. This may indicate a smeared first-order transition. Note, that in CeCoIn$_5$, a sharp and large peak of the specific heat is found at low-temperatures, which has proven to be due to a first-order transition.

In Figure 2, we compare the zero- and high-field SC transition anomalies of Ce$_2$PdIn$_8$ and CeCoIn$_5$ (Ref. 27) on a reduced temperature scale. At $H = 1.8$ T for the former and 11 T for the latter system, respectively, $T_c$ is suppressed to 20% of its zero-field value. The peak at the SC to normal transition in the latter system is much sharper indicating a much stronger first-order nature of SC transition in CeCoIn$_5$. The peak height for CeCoIn$_5$ has been reported to be very sensitive to impurity scattering. Only 0.22% of Cd-doping suppresses the sharp peak in specific heat significantly and leads to a very similar C/T dependence as found for 1.8 T in Ce$_2$PdIn$_8$. We may thus associate the reduced peak height for Ce$_2$PdIn$_8$ to the about ten-times shorter electronic mean free path, compared to undoped CeCoIn$_5$ (4000 Å). The HFLT phase in undoped CeCoIn$_5$ causes an additional jump in $C_{el}(T)/T$ of about $\sim 0.2$ J/mol·K$^2$ at 11 T, whereas in Ce$_2$PdIn$_8$ no anomaly could be detected within the measurement error of $\sim 0.02$ J/mol·K$^2$ at 0.1 K.

Measurements of the electronic specific heat coefficient as a function of magnetic field, displayed in Figure 3, also show no sign of an anomaly in addition to $H_{c2}$. Figure 4 shows the phase diagram of Ce$_2$PdIn$_8$ for two perpendicular magnetic field directions, revealing an almost isotropic field effect on superconductivity. The strongly convex shape of $H_{c2}(T)$ indicates a pronounced effect of Pauli-limiting, dominating over orbital limiting. In CeCoIn$_5$, $H_{c2}$ is also strongly Pauli limited and furthermore displays a pronounced anisotropy, which could...
be related to the anisotropic normal state spin susceptibility. For CePdIn₈, by contrast, the upper critical field is almost isotropic, in consistent with the much weaker anisotropy of the reported normal-state susceptibility.

Next, we turn to the signatures of NFL behavior in the normal state. As shown in Figure 5, for fields slightly above $H_{c2}$, a logarithmic divergence of the specific heat coefficient is found over more than one decade in $T$. The entropy release in zero field at $T_c = 0.68$ K due to the formation of the SC state is almost fully balanced at $H = 2$ T (cf. inset of Fig. 1a), indicating that the same degrees of freedom are responsible both for the NFL behavior and the SC state. At $H > H_{c2}$ for both magnetic field directions, Fermi liquid behavior is gradually recovered, as seen from the evolution towards a temperature independent specific heat coefficient $C_{el}/T$ in the insets of Fig. 5, we plot the magnetic field dependence of the Sommerfeld coefficient, $\gamma(H)$, obtained from the saturated $C_{el}/T$ values. Upon reducing the field from large values down to $H_{c2}$, divergent behavior is found in $\gamma(H)$. This indicates the existence of a field-induced QCP in close vicinity to the upper critical field of superconductivity, similar as in CeCoIn₅. In order to obtain information on the position of the QCP, as well as on the nature of the underlying quantum critical fluctuations, we have carefully compared the data with the predictions of the Hertz-Millis theory for an AF QCP in either 2D or 3D. The predicted field dependences are $\gamma_0 + \gamma_h \ln[1/(H - H_c)]$ (2D) and $\gamma_0 - \gamma_h \sqrt{H - H_c}$ (3D), respectively. We have tried least squares fitting of our data using $\gamma_0$, $\gamma_h$ and $H_c$ as free parameters. The field-dependence expected for 3D critical fluctuations appears to be too weak to explain the experimental $\gamma$ values at sufficiently large fields. A much better description could be obtained over one decade in $h=H-H_c$, using the expectation for 2D critical behavior. The obtained quantum critical fields for the 2D description are $H_c = 2.0 \pm 0.2$ T and $1.7 \pm 0.3$ T for $H \perp$ and $||$ to [001], respectively. These critical fields are very close to $H_{c2}$. Note, that recent electrical resistivity measurements have also suggested a field-induced QCP very close to $H_{c2}$ in this system. Furthermore, at $H \approx H_{c2}$ both the linear temperature dependence of the electrical resistivity, as well as the logarithmic temperature dependence of the specific heat coefficient are in agreement with the theoretical prediction for a 2D AF QCP.

In summary, we have studied the effect of magnetic fields on the SC and normal-state behavior in CePdIn₈ by measuring the specific heat at low temperatures and high magnetic fields. Even though the upper critical field $H_{c2}(T)$ is strongly Pauli-limited, a phase transition into a HFLT phase as seen in the relative CeCoIn₅ has not been detected. It has been recently reported that an extremely small amount of impurities destroys (or smears out) this
phase transition in CeCoIn$_5$. For 0.05% Cd-doped or 0.08% Sn-doped CeCoIn$_5$ it could not be detected by specific heat measurements any more.\cite{10,11} It has also been shown theoretically that a very small amount of impurities smears out the FFLO phase.\cite{12} Although Ce$_2$PdIn$_8$ is a clean limit superconductor, the electronic mean free path of the investigated single crystal is about ten times smaller compared to CeCoIn$_5$. This seems reflected in the much larger residual specific heat coefficient $C_2/T$ at zero field and reduced peak height of the SC transition in high fields. From the comparison of the respective specific heat data, we find the investigated Ce$_2$PdIn$_8$ single crystals comparable to 0.22% Cd-doped CeCoIn$_5$.\cite{13} Thus, a possible HFLT transition in Ce$_2$PdIn$_8$ could already be destroyed or smeared out by impurity scattering. Furthermore, we note that the upper critical field of the latter compound is almost isotropic, whereas it displays a pronounced anisotropy in CeCoIn$_5$. The observed NFL normal-state behavior in Ce$_2$PdIn$_8$ appears to be rather similar as found in CeCoIn$_5$. We have obtained a logarithmic divergence of $C_2(T)/T$ at fields near $H_{c2}$ and a logarithmic field dependence of the Sommerfeld coefficient, which both are compatible with 2D AF fluctuations due to a QCP near $H_{c2}$, in close similarity to CeCoIn$_5$. This work has been supported by the German Science Foundation through FOR 960 (Quantum phase transitions).

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