Pressure Dependence of the Fulde-Ferrell-Larkin-Ovchinnikov State in $CeCoIn_5$

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Pressure studies of the thermodynamics of $CeCoIn_5$ under magnetic fields $H \parallel c$ and $H \parallel ab$ have been made up to $P = 1.34$ GPa. We recorded the signature of the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state for all pressures when $H \parallel ab$. Also remarkably, the FFLO regime suddenly expands for $P = 1.34$ GPa. With the help of a microscopic theory for d-wave superconductivity, we have extracted the gyromagnetic ratio g and the Fermi velocities v_a and v_c. Our study is the first evidence for the existence of the FFLO state away from the influence of the antiferromagnetic fluctuations. We find a close parallel between the T-P phase diagram of $CeCoIn_5$ and the T-x phase diagram of the high-T_c cuprates, where x is the hole concentration.

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As is well known, type II superconductivity (SC) in the presence of a magnetic field is suppressed by two main mechanisms: the orbital effect [1] and/or the Pauli effect (i.e., the spin polarization [2,3]). If orbital pair breaking is negligible relative to the Pauli limiting effect, a spatially periodic modulation of the superconducting order parameter $\Delta(\mathbf{r})$ will take place for $T \leq 0.55T_c$ in sufficiently clean systems. This new periodic state (the FFLO state) was predicted 40 years ago by Fulde and Ferrell [4] and Larkin and Ovchinnikov [5]. However, the realization of such a state in conventional s-wave superconductors is almost impossible [6]. Gruenberg and Gunther [7] showed that the FFLO state is feasible even in the presence of the orbital term and their ansatz plays a crucial role, since it is also valid for d-wave or other nodal superconductors.

The appearance of unconventional superconductors such as the heavy-fermion, organic, and high-T_c compounds opens up a new window for exploration of the FFLO state. Many of these materials have a layered structure with larger Fermi velocities within the conducting plane. This reduced electronic dimensionality provides a path to substantially reduce the orbital limiting effect providing magnetic field is applied in a geometry (parallel to the conducting plane) such that the orbital currents would have to flow in prohibited directions. Technical advances in single crystal growth allows one to obtain samples with an electronic mean-free path of the order of microns that favors the FFLO state. Finally, for d-wave SC the FFLO state has a more extended stability region and is more robust against impurities [8] than for the s-wave superconductors [9].

Recently a new candidate for the realization of the FFLO phase has been found in $CeCoIn_5$. $CeCoIn_5$ exhibits the highest $T_c \approx 2.3$ K among the Ce-based heavy-fermion superconductors [10] and has been established as a $d_{x^2-y^2}$-wave superconductor [11,12] similar to the high-T_c cuprates. At ambient pressure $CeCoIn_5$ fulfills all the necessary conditions for the existence of the FFLO state. First of all, the system is in the extremely clean limit (i.e., $\ell_{tr}/\xi_0 = 14$ [13] where ℓ_{tr} is the quasiparticle mean-free path and ξ_0 the coherence length). Further, the Maki parameter $\alpha = \sqrt{2}H_{orb}/H_P$ [14], where H_{orb} is the orbital limiting field and H_P is the Pauli limiting field, takes the value 5.8 for $H \parallel ab$, which is significantly larger than the minimum value ($\alpha = 1.8$) required for s-wave superconductors [7]. Indeed, there are several reports providing strong evidence for the realization of the FFLO state in $CeCoIn_5$ and/or a first order phase transition (PT) to the superconducting state for $T \leq 0.7$ K [15–21].

Although no magnetic order is evident, resistivity and specific heat show non-Fermi liquid (NFL) behavior persisting over an extended temperature range in the normal state [10,22]. Moreover, strong spin fluctuations above T_c are revealed by nuclear quadrupole resonance (NQR) and NMR experiments [23]. All these facts are corroborated by a comparison to the closely related antiferromagnet $CeRhIn_5$ ($T_N = 3.8$ K) [24] and suggest that $CeCoIn_5$ is in proximity to a magnetic quantum critical point (QCP) situated at a slightly negative pressure. Detailed studies of resistivity and specific heat in applied magnetic field point also to a field-induced QCP existing in the vicinity of the upper critical field $H_{c2}(0)$ for the field in the basal plane [25,26] as well as for the field perpendicular to this plane [27]. Here, it is assumed that the ordered phase, possibly antiferromagnetic order, is superseded by SC. Recently it was proposed that the NFL regime in the phase diagram is replaced by an unconventional d-density wave phase in agreement with the giant Nernst effect [28,29] and angle dependent magnetoresistance results [30].

In this Letter we present specific heat studies on $CeCoIn_5$ under applied pressure and in high magnetic fields that reveal the pressure evolution of the low temperature anomaly ascribed to the FFLO state. In order to rule out the magnetic QCP related origin of this anomaly, one has to drive the system away from the influence of magnetic fluctuations. This is accomplished by applying

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hydrostatic pressure: $CeCoIn_5$ moves away from quantum criticality and enters a Fermi liquid behavior at high pressure [31–34]. Therefore, pressure studies are an ideal tool to investigate the origin of the second anomaly in the SC state.

We used a newly developed miniature piston-cylinder type pressure cell (machined completely from CuBe) for specific heat (C) measurements on high quality single crystals of $CeCoIn_5$. The experiments were carried out for magnetic field applied parallel to the tetragonal ab plane as well as parallel to the c axis in a dilution cryostat ($T \gtrsim 100$ mK, $H \leq 12$ T) and in a ^{3}He cryostat ($T \gtrsim 350$ mK, $H \leq 14$ T). The pressure cell allowed precise orientation of the crystals with respect to the magnetic field. Pb and a small amount of Apiezon-N grease served as the pressure transmitting medium. The inductively measured shift of the SC transition temperature of Pb under pressure served as a pressure gauge. It is important to note that the width of the SC transition of lead does not increase with increasing pressure. A comparison of the specific heat data at zero field, taken in the miniature pressure cell, to previous measurements, where a liquid pressure medium was utilized, confirms the quasihydrostatic conditions of the recent setup. The addenda, including the contribution of the cell and the pressure transmitting medium, was carefully measured and subtracted from the raw data for each magnetic field.

The specific heat data were collected for field $H \parallel ab$ and $H \parallel c$ at $P = 0$, 0.45, and 1.34 GPa. In the following, we focus on data obtained with magnetic field in the basal plane, where the possible existence of the FFLO state has recently attracted much attention. T_c increases from $T_c(0\ \text{GPa}) = 2.242$ K to $T_c(0.45\ \text{GPa}) = 2.428$ K and to $T_c(1.34\ \text{GPa}) = 2.584$ K, in good agreement with T_c determined from resistivity data [35] and previous specific heat measurements [32] under pressure. Figure 1 shows the specific heat data taken at $P = 0.45$ GPa in a series of magnetic fields. At low magnetic fields the specific heat at the SC transition reveals a mean-field-like shape. With

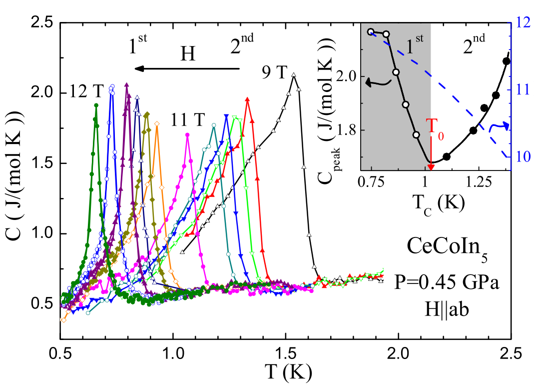

FIG. 1 (color online). $C(H, T)$ vs T at $P = 0.43$ GPa. The SC transition evolves from second to first order with increasing H. Inset: C_{peak} (C at the maximum of the SC anomaly) on the left axis and $H_{c2}(T)$ (dashed line) on the right as a function of T.

increasing field the anomaly sharpens, despite the PT line $H_{c2}(T)$ being crossed at a glancing angle at high magnetic fields. In the case of a second order PT one expects a significant broadening of the PT in this field range; in contrast, the peak becomes more symmetrical, indicating the change from a second to a first order PT at the magnetic field H_0 corresponding to $T_c = T_0$. Additionally, the height of the anomaly shows a minimum at T_0, typically observed at the crossover from a second to a first order PT (inset of Fig. 1). The specific heat results, together with magnetization data [36], provide strong evidence that the PT to the SC state at high magnetic fields at 0.45 and 1.34 GPa (data not shown) is first order as it is at atmospheric pressure. The crossover temperature T_0 increases from $T_0(0\ \text{GPa}) = (0.875 \pm 0.05)\text{K} = 0.39T_c$ to $T_0(0.45\ \text{GPa}) = (1.025 \pm 0.1)\text{K} = 0.42T_c$ and further to $T_0(1.43\ \text{GPa}) = (1.170 \pm 0.1)\text{K} = 0.44T_c$. The corresponding crossover fields are $H_0(0\ \text{GPa}) = 10.5$ T, $H_0(0.45\ \text{GPa}) = 11.2$ T, and $H_0(1.34\ \text{GPa}) = 12.4$ T. Magnetization studies find a similar T_0 for $H \parallel c$ at a fixed pressure [36], which we are not able to resolve unambiguously in our specific heat data.

The large nuclear Schottky contribution, C_{Schottky}, coming from both pressure cell and sample, causes a relatively high scattering in the low temperature specific heat $\Delta C = C - C_{\text{Schottky}}$. Therefore, we performed at least two temperature runs for each field in order to avoid any artifacts in the data. The electronic specific heat ΔC in high magnetic fields exhibits a second anomaly inside the SC state at low-T (which we denote T_{FFLO}) at ambient pressure, visible even when the sample is measured inside the pressure cell. The $T_{\text{FFLO}}(H)$ dependency we find for $P = 0$ GPa is in very good agreement with [15]. This feature can be traced as well under pressure, at $P = 0.45$ GPa and $P = 1.34$ GPa (Fig. 2). The tiny anomaly is comparable in size to the one found at ambient pressure. $C(T)$ at $P = 0.45$ GPa (Fig. 2, upper panel) shows the evolution of T_{FFLO} with magnetic field. For this pressure, at 10.4 T the transition from the normal to the SC state is still of second order, but a second anomaly is already observable at low-T. Only for $H > 11.2$ T (11.5 and 12 T depicted in Fig. 2) is the SC transition first order. This behavior is different from results at ambient pressure. Here, at the field where the second anomaly is found, the transition from the normal to the SC state is already of first order. We obtain similar results for $P = 1.34$ GPa, where, at fixed field, a normal to SC state PT, of a second order, is followed by a second anomaly in $C(T)$.

In the following we discuss the evolution of the H-T phase diagram with pressure and show that the necessary prerequisites required for the FFLO state are still satisfied. At ambient pressure we have established a H-T phase diagram for $CeCoIn_5$ as was previously observed [16,17]. Figure 3 displays the H-T phase diagram for $P = 0$, 0.45, and 1.34 GPa for $H \parallel ab$ and for $H \parallel c$. First we note the increasing anisotropy between $H_{c2}(0) \parallel ab$ and $H_{c2}(0) \parallel c$ upon applying pressure. The anomaly at T_{FFLO} appears first

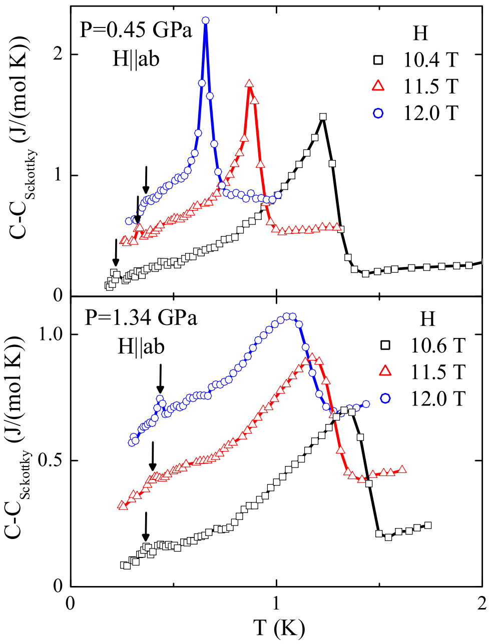

FIG. 2 (color online). Electronic contribution of low temperature specific heat $C - C_{\mathrm{Schottky}}$ for selected magnetic fields H close to $H_{c2}(0)$ at $P = 0.45$ GPa (upper panel) and $P = 1.34$ GPa (lower panel). Data at 11.5 T (△) and 12 T (○) are shifted by 0.25 J/(mol K) and 0.5 J/(mol K), respectively. The arrows indicate T_{FFLO}.

at almost the same magnetic field (≈10.5 T) independent of pressure. With increasing field $T_{\mathrm{FFLO}}(H)$ shifts to higher temperatures for all pressures, but is almost unaffected by a applying only a small pressure. Increasing the pressure to $P = 1.34$ GPa, however, moves $T_{\mathrm{FFLO}}(H)$ to higher temperatures, and $H_{c2}(0)$ increases considerably. Under finite pressure, with our setup, we are not able to follow $T_{\mathrm{FFLO}}(H)$ all the way to the tricritical point in the phase diagram, because at lowest temperatures our maximum magnetic field is limited to 12 T. Like $T_{\mathrm{FFLO}}(H)$ at 0 and

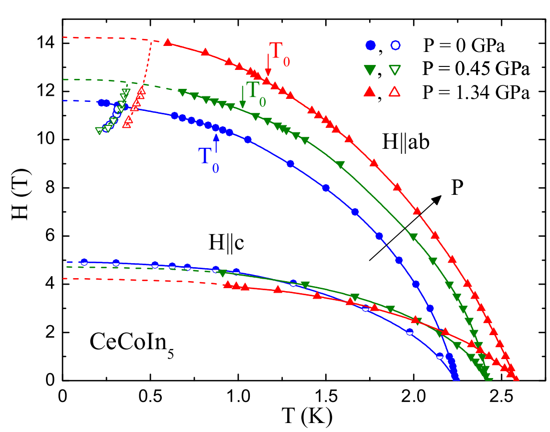

FIG. 3 (color online). H-T phase diagram for magnetic field $H \parallel c$ and $H \parallel ab$. Arrows mark the crossover temperature T_0 from a second to a first order PT into the SC state. Closed (half-filled symbols, data taken from [15]) symbols indicate $H_{c2}(T, P)$ and open symbols the second anomaly inside the SC phase.

0.45 GPa, the initial slope of the upper critical field $H'_{c2} = dH_{c2}/dT|_{T=T_c}$ of -30.5 T/K at $P = 0$ GPa and -29.4 T/K at 0.45 GPa also remains roughly the same. Correspondingly, the orbital critical field $H_{\mathrm{orb}} = 0.7T_c|H'_{c2}|$ [37] does not change significantly. We obtain $H_{\mathrm{orb}}(0\ \mathrm{GPa}) = 47.9$ T and $H_{\mathrm{orb}}(0.45\ \mathrm{GPa}) = 49.9$ T. However, at $P = 1.34$ GPa, H'_{c2} decreases drastically, by nearly one-half to $H'_{c2} = -16.4$ T/K, and consequently $H_{\mathrm{orb}}(1.34\ \mathrm{GPa}) = 29.7$ T. To determine the Maki parameter $\alpha = \sqrt{2}H_{\mathrm{orb}}/H_{\mathrm{P}}$, we need to estimate the Pauli limiting field H_{P}. It is reasonable to assume $H_{\mathrm{P}} \approx H_{c2}(0)$. With this estimation for H_{P} we get $\alpha(0\ \mathrm{GPa}) = 5.8$, $\alpha(0.45\ \mathrm{GPa}) = 5.7$, and $\alpha(1.34\ \mathrm{GPa}) = 2.9$, still sufficiently large compared to the required minimum value $\alpha = 1.8$ for s-wave superconductors [7] to realize the FFLO state. ℓ_{tr}/ξ_0 increases substantially with pressure for both field orientations. To estimate ξ_0 we used the relation $\xi_0 = \sqrt{\frac{\Phi_0}{2\pi H_{c2}(0)}}$ and followed the scheme of Orlando *et al.* [38] for ℓ_{tr} (see [39] for results). The increase of ℓ_{tr} with increasing pressure is essentially caused by the drop of the normal state resistance [31,35].

We have analyzed the $H_{c2}(T)$ data in Fig. 3 in terms of the theory [18] in which d-wave SC and the Gruenberg-Gunther ansatz [7] are incorporated within the weak-coupling BCS model. The effect of the quasiparticle mean-free path is neglected, which is valid for $CeCoIn_5$ since $\ell_{\mathrm{tr}}/\xi_0 \gg 1$. We can describe $H_{c2}(T)$ for both $H \parallel c$ and $H \parallel ab$ (we neglected the small in-plane anisotropy for $H \parallel ab$). The result is summarized in Table I. The Fermi velocities v_a and v_c are deduced from dH_{c2}/dT at $T = T_c$ for $H \parallel c$ and $H \parallel ab$, while the g value is obtained from $H_{c2}(T)$ for $H \parallel ab$ and $T < 0.7$ K. The $H_{c2}(0)$ values are theoretical predictions and are in fair agreement with the experimental data. The theoretical $H_{c2}(T)$ curves, plotted in Fig. 4, are obtained using T_c, v_a, v_c, and g as parameters and found to describe the experimental phase diagram quite well. The relevance of the g factor at low T is underscored by the considerable offset induced by only a slight change of the g value (dashed lines in Fig. 4). At $P = 1.34$ GPa a tiny decrease of the g factor for $T < T_0$ seems to describe the data better. However, such a weak $g(H)$ dependence is not accounted for by the current theory and a

TABLE I. Experimental values for T_c and estimation for α (see text for details). g factor, Fermi velocity v_a and v_c, and $H_{c2}(0)$ extracted from fitting of $H_{c2}(T)$.

P (GPa)	0	0.45	1.34
T_c (K)	2.242	2.428	2.584
α	5.8	5.7	2.9
g	0.632	0.6365	0.554
v_a (m/s)	7.47×10^3	7.89×10^3	9.59×10^3
v_c (m/s)	3.39×10^3	3.37×10^3	3.60×10^3
$H_{c2}(0)$ (T)	11.73	12.72	14.28

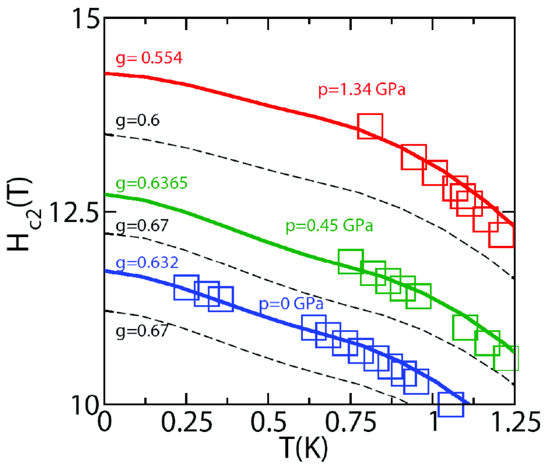

FIG. 4 (color online). Theoretical fits to upper critical field data at different pressures. See text for details.

different theoretical approach using, e.g., a generalized Fermi liquid theory might be required.

In conclusion, we have studied the pressure evolution of the superconducting H-T phase diagram of $CeCoIn_5$ by specific heat measurements. The FFLO anomaly inside the SC state is present at all pressures, reinforcing the existence of the FFLO state at low-T close to $H_{c2}(0)$. Our result is the first evidence of the existence of this state away from the influence of the strong magnetic fluctuations clearly suggesting its genuine FFLO nature. The first order character of the SC PT at high magnetic fields persists under pressure and the FFLO region in the phase diagram expands upon reducing the spin fluctuations, consistent with the model proposed in [40]. We have analyzed the $H_{c2}(T)$ data of $CeCoIn_5$ within the microscopic model which includes (a) the layered structure (or quasi-two dimensionality), (b) $d_{x^2-y^2}$-wave SC, and (c) the Gruenberg-Gunther ansatz. This weak-coupling model gives an excellent description of the observed $H_{c2}(T)$ curves for both $H \parallel c$ and $H \parallel ab$. Table I reveals the pronounced changes in g and the Fermi velocity in the basal plane v_a between $P = 0.45$ and 1.34 GPa. The reduction of the effective mass m^* and the rise of v_a with increasing P indicate a decrease of the quasiparticle-quasiparticle interaction while the simultaneous enhancement of T_c might be induced by an increase of the chemical potential [41]. The increase of v_a is in a qualitative agreement with the de Haas–van Alphen studies under pressure [33]. We recall that $P = 1.34$ GPa corresponds to the top of the SC dome in $CeCoIn_5$, in analogy to the top of the doping-induced dome in the high-T_c cuprates. There is thus an inescapable parallel between the high-T_c cuprates and $CeCoIn_5$ [41]. Indeed, the exploration of $CeCoIn_5$ from this point of view should be very fruitful.

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