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Interplay of magnetism and superconductivity in $CeCoIn_5$

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Abstract. $CeCoIn_5$ is a heavy fermion superconductor which appears to be straddling the boundary between the superconducting and magnetic ground states. At the superconducting critical field H_{c2} this material displays NFL behavior in transport and thermodynamic properties, pointing to a QCP at H_{c2}, and hinting at the presence of magnetic fluctuations, probably due to an AFM order superseded by the superconductivity. In the High-Field-Low-Temperature (HFLT) corner of the superconducting phase of $CeCoIn_5$, within 20% off H_{c2}, an additional phase appears within the superconducting phase, and the normal-to-superconducting transition itself becomes first order. This behavior is consistent with a strong Pauli limited superconductivity, and the low temperature high field phase being an inhomogeneous superconducting FFLO phase. Recent NMR experiments, however, point to a distribution of magnetic field on the scale of the crystallographic unit cell, and not on the scale of the superconducting coherence length expected of an FFLO order. Experiments on $CeRhIn_5$ under pressure show magnetic field induced AFM order within the superconducting phase, with some similarities to the phase diagram of $CeCoIn_5$. Could the HFLT phase transition be due to magnetic order? We need a picture of a magnetism "attracted" to superconductivity to explain the data on the HFLT phase in $CeCoIn_5$.

1.1 Introduction

$CeCoIn_5$ is a heavy fermion with a record high (for heavy fermion compounds) superconducting transition temperature $T_c = 2.3K$ [1]. $CeCoIn_5$ forms in a tetragonal crystal structure which can be thought of layers of $CeIn_3$ (derived from the parent cubic compound $CeIn_3$) separated by the layers of $CoIn_2$ along the c-axis, as shown in Fig. 1.1.

This perspective on the crystal structure of $CeCoIn_5$ as being quasi-two-dimensional is supported by both band structure calculations [2] and deHaas-vanAlphen measurements [3–5], which uncovered large undulating cylindrical parts of the Fermi surface with the axis along the [001] direction. The crystalline anisotropy leads to anisotropy in physical properties, including

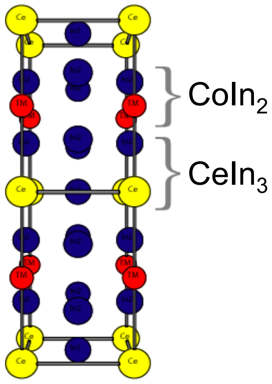

Fig. 1.1. Crystal structure of $CeCoIn_5$, showing the layers of $CeIn_3$ and $CoIn_2$.

magnetization (about a factor of two, with an easy axis along [001]). The superconducting critical field H_{c2} is itself anisotropic, with $H_{c2} = 4.95$ T for $H \parallel$[001] and 11.6 T for $H \parallel$ [100] [6].

$CeCoIn_5$ exhibits a number of fascinating properties in the vicinity of H_{c2}. Figure 1.2 shows the phase diagram of $CeCoIn_5$ with field $H \parallel$ [110], and introduces the main subjects of this paper. These include a Quantum Critical Point (QCP), coinciding with the superconducting critical field H_{c2}, and a

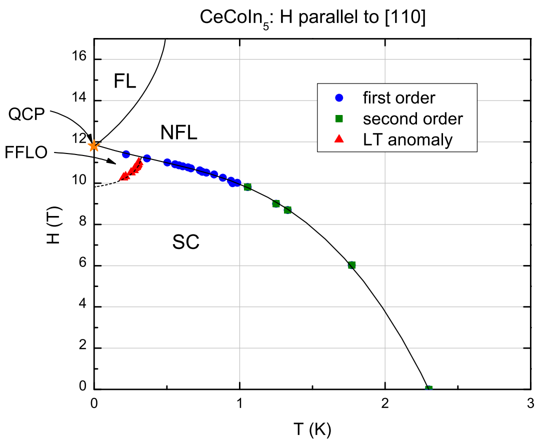

Fig. 1.2. Phase diagram of $CeCoIn_5$ for $H \parallel$[110]. SC transition changes from second to first order at $\approx$ 1 K and 10 T. A possibly FFLO state occupies the high field - low temperature corner of the superconducting phase, and a Quantum Critical Point (QCP) lies close to the superconducting critical field H_{c2}.

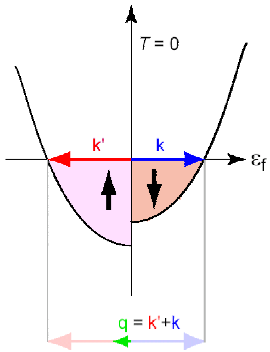

Fig. 1.3. Schematic of the formation of the FFLO state and Pauli limiting of the BCS state.

purported FFLO state (see below) in the high field-low temperature corner of the superconducting phase (the HFLT state). It is the interplay of these two phenomena and underlying interactions that is the main question that we are attempting to address below.

The subject that received the most attention since its discovery [7] is the novel HFLT phase which exists below 300 mK and between 10 T and H_{c2} = 11.6 T, with the field applied within the basal plane. It was suggested that this phase might be a realization of the spatially inhomogeneous Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) superconducting state, named after its discoverers (theoretically) over forty years ago [8, 9].

The FFLO phase is a result of a superconducting condensate minimizing the Zeeman energy of electron spins in magnetic field. Figure 1.3 gives a schematic representation of the effect of the magnetic field on both superconducting and normal states via Zeeman splitting of the spin up and spin down electron bands. In a spin-singlet BCS superconductor electrons form cooper pairs with opposite electron spins. As a result, the two electrons' Zeeman energies cancel each other. In the normal state electron spins would preferentially point along the magnetic field, lowering total energy of the system and leading to Pauli susceptibility. The resulting competition between the superconducting condensate energy and the Pauli energy provides a mechanism for suppression of superconductivity at the Pauli limiting field H_P [10], in addition to the so-called orbital limiting (with characteristic orbital limiting field H^0_{c2}) due to the opposite forces by the magnetic field on the two electrons of the cooper pair.

There are several types of systems that are traditionally thought to be good candidates to display the Pauli limiting and the FFLO physics. They

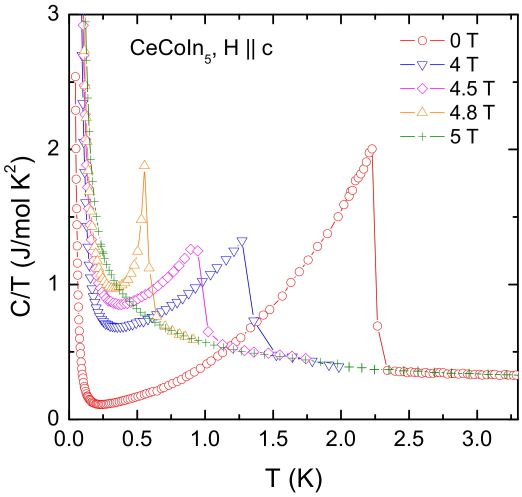

Fig. 1.4. Specific heat of $CeCoIn_5$, showing transition from second order character of the SC anomaly below 4.5 T to first order at 4.8 T.

are the systems that have weak or zero orbital limiting, and include superconducting films with magnetic field in the film's plane, low dimensional organic superconductors, and the heavy fermion materials. $CeCoIn_5$ is quasi-2D and a heavy fermion compound, and therefore is a good system in which to look for an FFLO state.

Phase diagram analysis allows us to estimate H_{c2}^0 (directly from the slope of the critical field at $T_c(H = 0)$) to range between 35 and 50 Tesla [6, 11], to be compared with the experimental superconducting critical field of $H_{c2} =$ 11.6 T. Large suppression of H_{c2} indicates that $CeCoIn_5$ is in a strong Pauli limit, i.e. Pauli limiting largely determines the observed H_{c2}.

The FFLO state takes advantage of the Zeeman energy of the superconducting electrons by pairing electrons at the Fermi energy with non-equal momenta k and k', leading to a cooper pair with a finite total momentum $q = k + k'$ (see Figure 1.3). The resulting superconducting state has an order parameter that is modulated in real space, e.g. $\Delta = \Delta_0 \cos(\boldsymbol{qr})$ for the Larkin-Ovchinnikov state.

It was predicted theoretically that in a strongly Pauli limited superconductor the order of the superconducting transition will change from second to first [12]. This effect indeed was observed in $CeCoIn_5$ for magnetic field both within the $a-b$ tetragonal plane and perpendicular to it [6,13]. Fig. 1.4 shows specific heat for $H \parallel [001]$, when $H_{c2} \approx 5$ T. For fields below 4.5 T the specific heat anomaly has a classic mean field shape, with a sharp step at T_c, followed

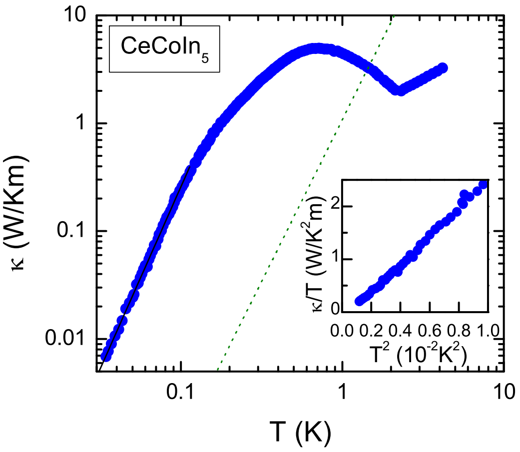

Fig. 1.5. Thermal conductivity of $CeCoIn_5$Ṗeak below T_c and T^3 behavior at low temperature indicate high purity of the sample and a long quasiparticle mean free path in the SC state.

by a gradual decrease at lower temperature. Magnetic field suppresses T_c and the size of the superconducting anomaly. The 4.8 T data, however, breaks that monotonic evolution: the anomaly narrows down, becomes symmetrical, and the peak hight increases compared to the data for 4.5 T. This behavior demonstrates that the order of the superconducting transition changes from second to first somewhere between 4.5 T and 4.8 T. Detailed analysis of additional data pinpointed the change to occur at $T_0 \approx 0.3T_c$ [13] for this field orientation. The first order superconducting transition in $CeCoIn_5$ was taken to be due to strong Pauli limiting effect, consistent with the phase diagram analysis above and in accord with theoretical expectations.

The next natural step was to look for the FFLO state in $CeCoIn_5$, since it should be driven by the same physics (Pauli limiting) that lead to the first order SC transition. The FFLO state itself is expected to be rather fragile in a sense that it should be easily destroyed by the minute amount of impurities. The last requirement for the formation of the FFLO state is therefore that the sample must be very clean, i.e. electron mean free path should be many times that of the superconducting coherence length. The first indications of the extreme purity of $CeCoIn_5$ came from the measurements of the thermal conductivity κ in $CeCoIn_5$ [14]. κ/T displays a very sharp kink at T_c, and rises dramatically by an order of magnitude with decreasing temperature, reaching a maximum at $T/T_c = 0.2$. This, combined with the fact that the number of heat carrying normal quasiparticles is drastically decreased between T_c

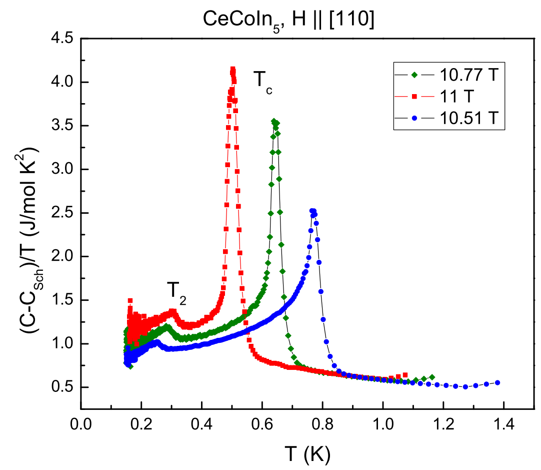

Fig. 1.6. Specific heat of $CeCoIn_5$ with $H \perp [001]$. Additional anomaly (marked as T_2) below a sharp first order transition into superconducting state at T_c indicates transition in to a potentially FFLO state.

and $0.2T_c$, lead to an estimate of the quasiparticle mean free path of several microns deep within the superconducting state, orders of magnitude greater than the superconducting coherence length (on the order of 100 Å).

Finally, to take advantage of the quasi-2D nature of $CeCoIn_5$ in the search for the FFLO state, specific heat measurements were performed with the field within the basal $a - b$ plane, where $H_{c2} = 11.6$ T. Some of the specific heat data that lead to the phase diagram in Figure 1.2 are displayed in Figure 1.6. In addition to a very sharp first order superconducting transition at T_c, additional anomaly within the superconducting state suggests transition into an FFLO state.

1.2 Magnetism in $CeCoIn_5$

In addition to the measurements described above, there has been a number of other reports in support of the FFLO scenario in $CeCoIn_5$, including a number of thermodynamic, transport, and microscopic studies. For a recent review, see Ref. [15]. However, recent NMR investigations [16] showed that there is a long range antiferromagnetic order within the HFLT phase. Figure 1.7 shows NMR spectra taken at 11.1 T for a range of temperatures from 890 mK to 50 mK. In(1) site lies in the Ce plane, whereas In(2) site lies outside of the Ce plane. The resonance associated with the In(1) site shifts to lower frequency, but remains sharp. The signal from the In(2) resonance disappears at the HFLT

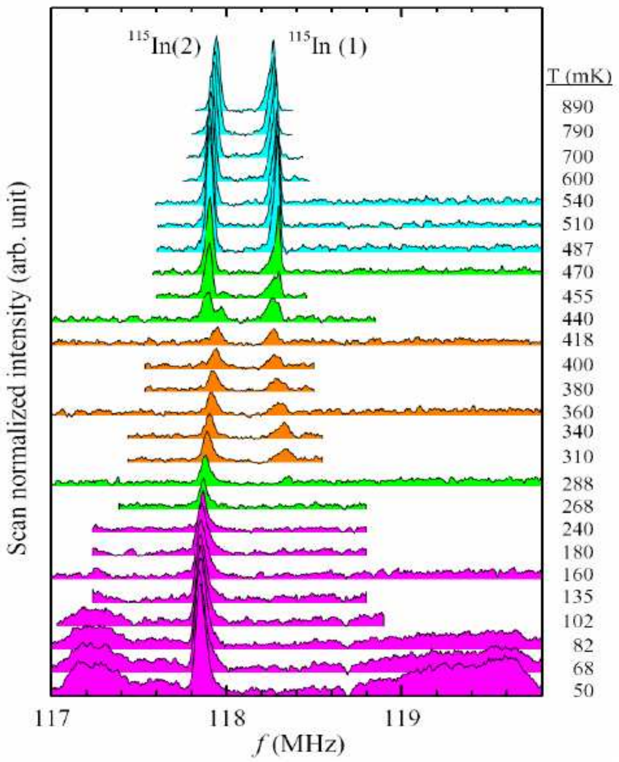

Fig. 1.7. NMR spectra of In(1) and In(2) transitions in $CeCoIn_5$ at 11.1 T. In (1) transition shifts down in frequency, whereas the In(2) shifts up in frequency.

transition at about 300 mK, and appears at much lower temperature below 100 mK as broad feature consisting of two two broad features, one peaked at 117 MHz and another at 119.6 MHz. This shape of the NMR resonance is characteristic of an incommensurate magnetic order. Therefore, the two In sites within a single unit cell experience different magnetic field distribution, which lead the authors to conclude that an AFM long range order is present in the HFLT state [16].

Is it possible that the HFLT phase is of magnetic origin? Previous studies [17, 18] have shown that there is indeed a nearby magnetic ground state, which manifests itself via non-Fermi-liquid (NFL) behavior in the normal state of $CeCoIn_5$. Figure 1.8 shows specific heat data of $CeCoIn_5$ for field $H \parallel [001]$ at the superconducting critical field $H_{c2} = 5$ T, and above it up to 9 T. Logarithmic divergence of $\gamma = C/T$ vs. T at H_{c2} is a non-Fermi-liquid behavior characteristic of the proximity to a Quantum Critical Point (QCP).

It was suggested [17] that the QCP is due to an AFM ground state that is superseded by superconductivity ($T_c > T_N$), and when the SC state is

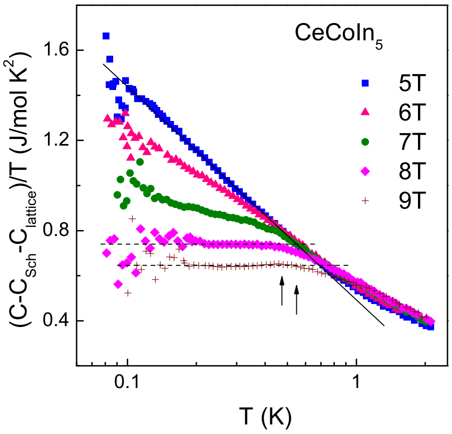

Fig. 1.8. Specific heat of $CeCoIn_5$ with H $\|$[001]. C/T diverges logarithmically at H_{c2}. Arrows indicate the onset of the Fermi liquid behavior (constant C/T) at higher fields.

formed, all of the Fermi Surface is gapped, leaving no states to participate in the AFM order. Such scenario leads to a phase diagram shown in Figure 1.9, where magnetic field suppresses superconductivity faster than magnetism, and T_c and T_N are driven to zero by roughly the same magnetic field ($H_{QCP} \approx H_{c2}$

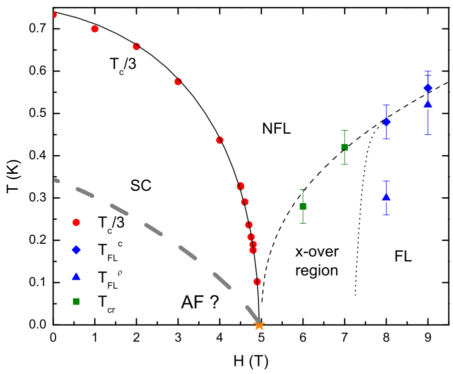

Fig. 1.9. Phase diagram of $CeCoIn_5$ with H $\|$[001], showing a large non-Fermi-liquid (NFL) region. Putative boundary of the avoided AFM state, which leads to the NFL behavior, is indicated by the dashed line.

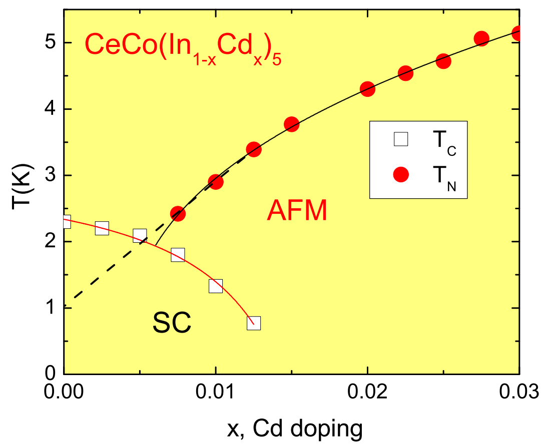

Fig. 1.10. Phase diagram of $CeCo(In_{1-x}Cd_x)_5$. Cd doping suppresses SC and stabilizes AFM state. Dashed line is a possible extrapolation of the AFM phase boundary to $x = 0$.

= 5 T). Several attempts were made to separate H_{c2} and H_{QCP} and uncover the AFM state. Doping Sn for In suppressed both T_c and T_N roughly equally, with the strongest $\log T$ divergence of γ remaining at H_{c2} [19]. Recent Cd-doping studies, however, finally revealed the underlying AFM state [20]. As Figure 1.10 shows, Cd doping of about 0.5% suppresses the superconducting state, and stabilizes the AFM ground state. The dashed line represents a possible extrapolation of the AFM phase boundary that would intercept the temperature axis at a positive value. $x = 0$ situation is in fact identical to the $H = 0$ one displayed in Figure 1.9. Thus, the AFM phase revealed by the Cd doping studies is the “avoided AFM phase” suggested to exist in $CeCoIn_5$ on the basis of earlier specific heat studies [17].

1.3 $CeRhIn_5$ under pressure: close relative to $CeCoIn_5$.

$CeRhIn_5$ is an ambient pressure antiferromagnet. Hydrostatic pressure suppresses AFM state and stabilizes superconductivity [21, 22]. Above the pressure $P_1 = 1.77$ GPa, $T_c > T_{AFM}$, and once superconducting state is stabilized, all the Fermi Surface is gapped, and AFM state does not develop below T_c. However, it was shown that application of magnetic field for pressure just above P_1 restores long range magnetic order [23]. Figure 1.11 shows reduced $H - T$ phase diagram of $CeRhIn_5$ for several pressures above P_1. Magnetic field induces an AFM state within the superconducting state, and the closer

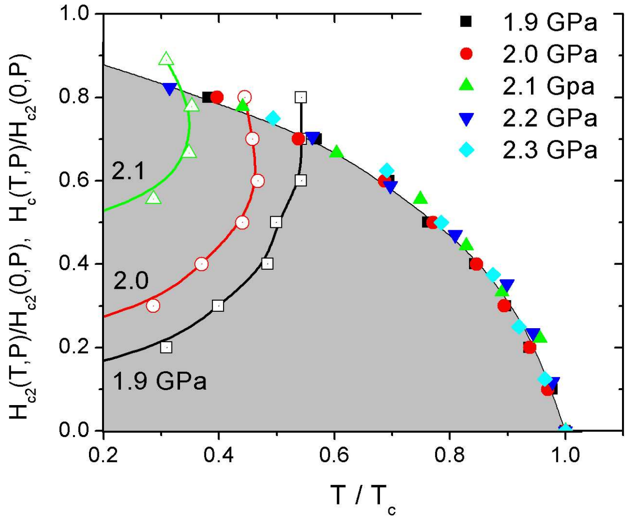

Fig. 1.11. Reduced phase diagram of $CeRhIn_5$ for several pressures $P > P_1$. When AFM ground state is close (. Dashed line is a possible extrapolation of the AFM phase boundary to $x = 0$.

the pressure is to P_1, the smaller magnetic field is required to induce the long range magnetic order. This phenomena was explained within the model [24] where AFM correlations set in within the vortex cores, and become long range order as the vortices come close together with increasing magnetic field. The shape of the field-induced AFM phases in $CeRhIn_5$ is reminiscent of that of the HFLT phase in $CeCoIn_5$, however, there is a very important difference. In case of $CeRhIn_5$ AFM state penetrates into the normal state above H_{c2}, whereas in $CeCoIn_5$ the boundary between the HFLT phase and the mixed superconducting vortex state does not cross the SC - normal phase boundary $H_{c2}(T)$. The HFLT phase is integrally connected to superconductivity, as if the AFM order is “attracted” to superconductivity, a unique situation.

1.4 HFLT phase and magnetism in $CeCoIn_5$ under pressure

As mentioned above, recent NMR studies of $CeCoIn_5$ found evidence of a long range AFM order within the HFLT phase. The question arises then whether the avoided AFM state, revealed by the Cd doping studies, is at the core of the HFLT phase? Does quantum criticality at H_{c2} lead to the formation of the HFLT state? Measurements under pressure give us important clues about the interplay of superconductivity and magnetism in $CeCoIn_5$.

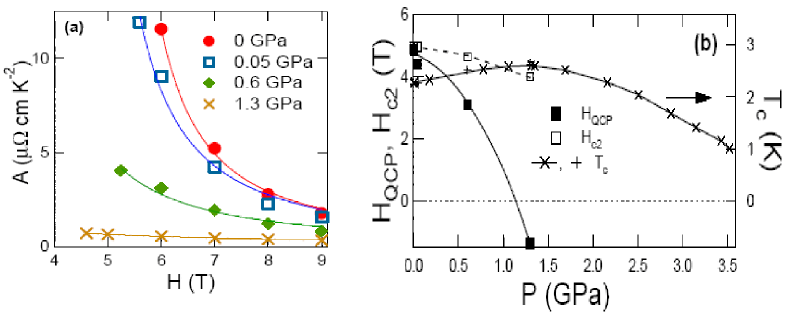

Fig. 1.12.

Specific heat measurements under pressure [11] revealed that pressure increases the extent of the superconducting phase, with both T_c and H_{c2} growing. In addition, the measurements showed that the phase space occupied by the HFLT phase itself increases as well. To test the connection between the QCP and the HFLT phase we investigated the dependence of the field H_{QCP} of the QCP on pressure [25] via the resistivity measurements.

Figure 1.12 shows the results of the analysis of the resistivity data for several pressures up to 1.3 GPa with field $H \parallel [001]$. For each pressure resistivity was fit to the expression $\rho = \rho_0 + AT^2$. The resulting values of the coefficient A of the quadratic term are shown in Figure 1.12(a). A is proportional to the quasiparticle mass, or, alternatively, to the density of states at the Fermi level. It is expected to diverge at a QCP, and we fit it to the expression $A = \alpha(H - H_{QCP})^\beta$. The resulting fits are displayed in the Figure 1.12(a). Figure 1.12(b) shows H_{QCP} obtained from the fit, together with H_{c2} and T_c, as a function of pressure. H_{QCP} is suppressed to zero by about 1.3 GPa, clearly separating from H_{c2}. This is a strong indication that the tendency toward magnetism is suppressed with pressure, while the HFLT state was shown to be enhanced by it. These results support the conjecture that the HFLT state is of non-magnetic, possibly FFLO, origin.

1.5 Conclusions

In this paper we presented results of several measurements that paint a picture of a complex state in $CeCoIn_5$ with subtle balance between tendencies toward magnetic order and superconductivity. As a result, the Quantum Critical Point due to the underlying antiferromagnetic state, coincides with the superconducting critical field in $CeCoIn_5$. In addition, a novel superconducting state develops in the high fied and low temperature (HFLT) corner of the state of the superconducting phase with the long range antiferromagnetic order. Regardless of whether the HFLT state is of purely magnetic origin, or magnetism accompanies a fundamentally FFLO state, we are presented here with magnetism that is stabilized in the superconducting state only, and does not extend into the normal state. This is a highly unique situation, exact opposite to the canonical picture of the competition between superconductivity and magnetism, and is worthy of detailed experimental and theoretical studies. Additional investigations of the HFLT state in $CeCoIn_5$ are required before a firm case can be made for its nature.

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