Anomalous Superconductivity and Field-Induced Magnetism in CeCoIn$_5$

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In the heavy fermion superconductor CeCoIn$_5$ ($T_c = 2.3$ K) the critical field is large, anisotropic and displays hysteresis. The magnitude of the critical-field anisotropy in the a-c plane can be as large as 70 kOe and depends on orientation. Critical field measurements in the (110) plane suggest 2D superconductivity, whereas conventional effective mass anisotropy is observed in the (100) plane. Two distinct field-induced magnetic phases are observed: $H_a$ appears deep in the superconducting phase, while $H_b$ intersects $H_c^2$ at $T=1.4$ K and extends well above $T_c$. These observations suggest the possible realization of a direct transition from ferromagnetism to Fulde-Ferrel-Larkin-Ovchinnikov superconductivity in CeCoIn$_5$.

The interaction of magnetism and superconductivity is a significant and long standing problem in condensed matter physics. Usually, the presence of magnetic order undermines superconductivity, but in heavy fermion materials, superconductivity and magnetism can coexist without deleterious consequences to the superconducting state. These systems provide an opportunity to explore the interaction of magnetic and superconducting order parameters as a function of temperature, pressure, or magnetic field. While antiferromagnetism interacting/coexisting with superconductivity is the case most often considered, examples of ferromagnetism coexisting with superconductivity have been reported recently. In heavy fermion superconductors the combination of large initial critical field vs. temperature slopes and long mean free paths also potentially allows for the observation of critical fields beyond the Pauli limit and, perhaps, inhomogeneous pairing states.

Recently the heavy fermion compound CeCoIn$_5$ was observed to superconduct at 2.3 K, the highest $T_c$ yet reported for a heavy fermion superconductor. Specific heat and thermal transport studies establish that the superconductivity in this material is unconventional and magnetically mediated. Because crystallographic anisotropy might play an important role in the properties of this tetragonal material and de Haas-van Alphen measurements reveal a two-dimensional character of the Fermi surface, a thorough investigation of the anisotropic critical field-temperature phase diagram was undertaken and is reported in this Letter. We observe not only an upper critical field, $H_c^2$, that varies differently as a function of angle in the (100) and (110) planes but also the existence of field-induced magnetic phases in both the normal and superconducting states of CeCoIn$_5$.

CeCoIn$_5$ forms in the tetragonal HoCoGa$_5$ crystal structure with lattice constants $a=4.62\AA$ and $c=7.56\AA$. The crystal structure consists of alternating layers of CeIn$_3$ and CoIn$_2$. The crystallographic axes of the flux-grown single crystals used in our experiments were determined by Laue x-ray diffraction. The [001] axis was parallel to the shortest dimension of the crystal and [100] and [010] axes were parallel to the natural edges of the nominally square crystals. The superconducting-normal phase boundary and the magnetization of CeCoIn$_5$ were determined by electrical transport, AC susceptibility and cantilever magnetometry measurements as a function of magnetic field ($0-200$ kOe) and temperature ($0.02-27$ K). Angular variations were measured using a rotating sample stage in a top-loading dilution refrigerator and in a $^3$He cryostat. Three different single crystals were studied, with consistent agreement among their measured $H_c^2$ values.

Fig. 1 shows a signal proportional to $\vec{M}$ plotted against applied magnetic field for both increasing and decreasing fields. The two traces are for the field applied along the [110] and [001] crystal axes at $T = 20$ mK. With increasing field, a narrow superconducting-normal transition, $\Delta H_c^2 < 10$ Oe, is clearly seen in the $\vec{H} \parallel [110]$ trace, and a somewhat broader transition is seen in the $\vec{H} \parallel [001]$ trace. These traces are typical of data used to construct the phase diagrams reported below. At $T=20$ mK and $\vec{H} \parallel [001]$, $H_c^2 = 50.5$ kOe and for $\vec{H} \parallel [110]$, $H_c^2 = 119$ kOe. Resistivity measurements (not shown) confirm that these transitions correspond to superconducting-normal transitions. For a material having $T_c = 2.3$ K, these values of $H_c^2$ are quite large: a simple estimate of the Clogston limit gives $H_c^2(T=0) = (18.6 \text{ kOe/K})T_c = 43 \text{ kOe}$.

For field applied along [110] the normal-superconducting resistive transition occurs at the same field on both up-
sweep and down sweep; however, the magnetization transition occurs at a lower field on the down sweep, suggesting an additional phase transition in the superconducting state. No such second transition is observed for H || [001].

An additional feature apparent in Fig. 1 is the peak in magnetization observed for H || [001] at $H_a$=28 kOe. Preliminary investigations show that this feature appears only below 100 mK and exhibits a complex dependence on field orientation and sweep direction. Although it will be discussed in detail elsewhere [13], we note here that $H_a$ appears to merge with $H_{c2}$ (up sweep) when the applied field is within 5 degrees of [110].

The angular dependence of $H_{c2}$ at 20 mK is shown in Fig. 2. The evolution of $H_{c2}$ for rotation of $\vec{H}$ from [001] into [100] is well described by the anisotropic effective mass model [14], taking $H_{c2}(\theta)$ as the up sweep value:

$$H_{c2}(\theta) = H_{c2}(\theta = 0) /[\cos^2(\theta) + \alpha \sin^2(\theta)]^{1/2} \quad (1)$$

where $\theta$ is the angle of the applied field out of the tetragonal basal plane and $\alpha$ is the ratio of effective masses $m^*(\theta = 0)/m^*(\theta = 90)$. The large value of $\alpha$=6.1 confirms the significant electronic anisotropy in CeCoIn$_5$ deduced from de Haas van Alphen measurements [15].

Rotating $\vec{H}$ from [001] into [110] produces a much more cusp-like angular dependence than Eq. 1 would predict. In this case, the data are well described by Tinkham’s equation for $H_{c2}$ as a function of angle in thin film superconductors [15]:

$$|H_{c2}(\theta)\sin(\theta)/H_{c2}(90)| + [H_{c2}(\theta)\cos(\theta)/H_{c2}(0)]^2 = 1. \quad (2)$$

Both sets of data in Fig. 2 were obtained using the same single crystal, so neither sample-to-sample variation nor demagnetization corrections can explain the different angular variations in $H_{c2}$. We also note that $H_{c2}(110)$=119 kOe while $H_{c2}(100)$=118 kOe which implies the existence of in-plane anisotropy in CeCoIn$_5$ (Fig. 1).

The angular dependence of $H_{c2}$ observed in the (110) plane is reminiscent of behaviors in granular thin film [17,18] and multilayer [19] systems. In fact the quality of the fit to our data is comparable to and extends over a wider angular range than that in Al films [17]. Why 2D behavior in one particular plane would be observed in bulk CeCoIn$_5$ is not understood. Band structure calculations suggest that the density of states in the Mn$_2$ layer in CeMn$_5$ is quite low [20] and leads to the speculation that perhaps CeCoIn$_5$ may behave as a pseudo CeIn$_3$:CoIn$_2$ multilayer system. Even if such a speculation were shown to be relevant, why the phenomenon would manifest itself in [001]-[110] rotations but not [001]-[100] rotations is unclear; however, it might be related to an in-plane modulation of the superconducting gap function [7,9] or to anisotropic Fermi surface nesting [3,10].

The difference in field between the upsweep and down sweep transitions in magnetization is a strong function of crystallographic direction (Fig. 3). The difference increases as the field is rotated toward [110] and has a maximum value of 25 kOe. As will be discussed below, CeCoIn$_5$ displays a metamagnetic transition at high fields, and the presence of a static magnetization in the sample complicates the determination of the down sweep transition field. The values shown in Fig. 2 have been corrected to account for the magnetization, $\vec{M}$, in the sample that contributes to the internal magnetic field, $\vec{B}$ according to the relation: $\vec{B} = \vec{H} + \mu_0\vec{M}$. Using this relation, we corrected an offset in the measured down sweep transition field around [110] that was the result of moving from a magnetic normal state into a superconducting one. The maximum contribution of $\vec{M}$ is estimated to be $\mu_0\vec{M}$=15 kOe along [110]. The separation in transition fields as a function of angle for the [100] rotation is shown in Fig. 2 as well. The difference between the up and down sweeps in this case is almost negligible (resulting in symbols in the figure that essentially overlap). The maximum field separation along [100] is only 0.8 kOe, a factor of 31 less than the value of 25 kOe that is found along [110].

The evolution of these transitions in the [110] direction in CeCoIn$_5$ with temperature also is anomalous. An H-T phase diagram for H || [110] in CeCoIn$_5$ is shown in Fig. 4. Three characteristic temperature ranges can be identified (see Fig. 3 for representative data): I) For T < 1.4 K, field-separated transitions in magnetization are observed and the changes in magnetization at the transitions are step-like; II) for 1.4 K < T < 2.3 K, no evidence for the lower-field transition nor step in magnetization at $H_{c2}$ is observed, but for fields greater than $H_{c2}$, a normal-state metamagnetic transition, occurring at $H_a$, is seen; and III) for T > 2.3 K only the metamagnetic transition is observed.

In Region I the field separation between magnetization transitions decreases with increasing temperature and at 0.5 K, $H_{c2}(\theta)$ has the same relative angular dependence (not shown) as at 20 mK (Fig. 1). The zero resistance transition occurs (regardless of field sweep direction) at the higher up-sweep value of $H_{c2}$ deduced from magnetization, and a new ferromagnetic-like (because of the observed steps in magnetization) first-order transition in the superconducting state emerges below the resistively-determined $H_{c2}$. This state is found below 0.6$T_c$ at high fields and its appearance depends strongly on the orientation of field with respect to crystallographic axes. Taken together, these observations are consistent with a spatially inhomogeneous Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) state [6]. The fact that no signature of a BCS-FFLO transition prior to the superconducting-normal transition is observed in up sweep magnetization may suggest that this transition is hysteretic in field or that the FFLO state is only stabilized by the presence
of magnetic order. High-field heat capacity and neutron scattering measurements should be able to clarify this issue.

Although the FFLO state is rarely observed \[21,22\], CeCoIn\(_5\) satisfies the essential conditions for its existence \[23\]: it is in the clean limit \[7\], has a quasi-2D Fermi surface \[8,9\], and has an \(H_c^2\) much larger than the Clogston limit. The transition from the normal state to the FFLO state is from ferromagnetic to superconducting, which to the best of our knowledge, is unprecedented. Recent calculations of Zeeman effects in d-wave superconductors \[24\] (e.g., CeCoIn\(_5\) \[7,16\]) suggest that an increase in \(H_c^2\) and the appearance of another magnetic transition, perhaps related to \(H_a\), at lowest temperatures is a consequence of an FFLO state in such a superconductor. If we are not observing FFLO superconductivity in the [110] direction, then the finite jump in \(\vec{M}\) in the superconducting state implies the coexistence of superconductivity with a spin-polarized state, the field-sweep dependent continuation of \(H_a(T)\) into the mixed state.

The signature for \(H_c^2(T)\) intersects \(H_b(T)\) and the signature of the FFLO state vanishes at \(H=80\) kOe and \(T=1.4\) K. Because the magnetization change at \(H_c^2\) disappears above 1.4 K, we used transport measurements to follow \(H_c^2\) up to \(T_c(H=0)\) with no observable hysteresis nor second transitions present. The change in magnetization at \(H_a\) is approximately a factor of 2.5 less than at \(H_c^2\) (for \(T<1.4\) K) and appears to be second order as a function of temperature. The signature for \(H_a\) weakens as \(\vec{H}\) is rotated away from [110] and is completely absent for \(H\parallel[001]\), again illustrating the anisotropic magnetic behavior of this material even in the normal state. Given the extent to which its evolution is influenced by superconductivity without deleterious effects on \(H_c^2\), it is tempting to identify the paramagnetic-magnetic normal state transition with spin polarization of a sheet of Fermi surface; quantum oscillation measurements to test this hypothesis are in progress.

In summary, we find a remarkable \(H_c^2\) anisotropy in CeCoIn\(_5\) that is correlated with the presence of a magnetic transition in the superconducting state for \(H\parallel[110]\). These data can be described empirically in terms of 2D superconductivity and suggest the formation of an FFLO state. \(H_c^2\) anisotropy also exists within the (100) plane but is describable by anisotropic band structure effects, and does not present hysteresis. We also have observed two new magnetic phases in CeCoIn\(_5\), one occurring deep in the superconducting state at very low temperature (\(H_a\)) and the other (\(H_b\)) manifesting itself as a field-induced metamagnetic transition that persists to at least 25 K.

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![FIG. 1](image1.png) Magnetization loops for field applied along [110] and [001] at 20 mK in CeCoIn\(_5\). Arrows indicate direction of field sweep.

![FIG. 2](image2.png) \(H_c^2\) as a function of angle for CeCoIn\(_5\) for field rotations from [110] to [001] (•= upsweep, ◦= downsweep) and from [100] to [001] (▼= upsweep, ▽= downsweep). The data points for the second curve fall on top of one another thus the open triangles are not visible. See text for fit equations.
FIG. 3. Magnetization as a function of field ($\vec{H} \parallel [110]$) at 3 characteristic temperatures (0.020 K, 1.8 K, and 5 K) in CeCoIn$_5$. The noise in the 0.020 K resistance measurement is due to flux popping in the magnet at low fields. Note that the sharp transition in the magnetization in the 0.020 K panel occurs at the onset of superconductivity as displayed in the resistance measurement. Arrow indicates position of zero-resistance transition for 1.8 K panel.

FIG. 4. H-T diagram for CeCoIn$_5$ with $\vec{H}$ applied in the (110) direction (the inset emphasizes the high-temperature range of the main figure). Circles and squares denote magnetization transitions (■=up sweep, □=downsweep) and (●=up sweep, ▲=down sweep). Triangles indicate resistively determined $H_C^\parallel$. Measurements were made with three different systems and the resultant offset between circles and crosses is due to slight differences in crystal alignment. The diamonds denote the field-induced magnetic transition that appears at 1.4 K. I, II, and III indicate the three regions in the phase diagram discussed in the text.

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