Detection of high-field superconducting phase in CeCoIn$_5$ with magnetic susceptibility

S.D. Johnson and R.J. Zieve

Physics Department, University of California Davis, Davis, CA 95616

J.C. Cooley

Los Alamos National Laboratory, Los Alamos, NM 87545

(Dated: May 5, 2014)

We measure the ac susceptibility of single-crystal CeCoIn$_5$ in dc field parallel to the c axis and find further evidence for a high-field phase transition within the superconducting phase in this orientation. We apply up to 2.3 kbar uniaxial pressure along the c axis and discuss the pressure dependence of the high-field phase. We also report the behavior of $H_{c2}$ under uniaxial pressure for field along c.

The heavy-fermion superconductor CeCoIn$_5$ and its isostructural cousins Ce(Ir,Rh)In$_5$ and Pu(Co,Rh)Ga$_5$ exhibit several intriguing features. Collectively these compounds have ground states that are superconducting, antiferromagnetic, or both. The materials have significant two-dimensional character, suggested by their layered crystal structure and confirmed through electronic structure calculations. Furthermore, within each family $T_c$ varies linearly with the ratio of lattice constants $c/a$, illustrating how closely geometry and superconductivity can be linked [1]. With a c-axis magnetic field CeCoIn$_5$ also has a quantum critical point near the upper critical field, which can be shifted into the superconducting phase with hydrostatic pressure [2].

Yet another unusual feature appears in the high-field, low-temperature portion of the superconducting regime. This has been studied mainly for magnetic fields applied in the basal plane, where $H_{c2}$ is about 11.8 Tesla and a signature within the superconducting phase is seen near 10 Tesla with magnetization [3], specific heat [4], neutron diffraction [5], and NMR [6] measurements. The additional signature moves to higher fields as temperature increases, intersecting the $H_{c2}(T)$ curve near 300 mK. These data are of particular interest since CeCoIn$_5$ is a good candidate to support the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) superconducting phase described almost 50 years ago [7, 8]. This state is marked by a non-zero quasiparticle center-of-mass momentum and a spatially periodic superconducting order parameter. CeCoIn$_5$ is in the extremely clean limit and has strong Pauli limiting of its critical field. In addition, the high-field phase (HFP) is accompanied by a change of the normal-superconducting transition from second-order at low fields to first-order at high fields, in agreement with FFLO theory [3-5, 10]. While the exact nature of the phase and its identification with FFLO remain in question [3-5, 10], the experimental evidence for the HFP with in-plane field is clear.

Much less effort has been devoted to CeCoIn$_5$ with magnetic field along the c axis. A few measurements have indicated a high-field phase for this orientation too, as seen both by signatures within the superconducting phase [11-13] and by a change of the normal-superconducting transition from second-order to first-order similar to that observed for in-plane field. However, other experiments have found no signatures [4, 9], leaving even the experimental situation unclear. In this report we provide further evidence for the HFP phase in CeCoIn$_5$ for $H||c$, along with the uniaxial pressure dependence of the HFP boundary up to 2.32 kbar.

We measured three samples oriented with uniaxial pressure, dc field, and ac field parallel to the c axis. The pressure is applied with a bellows setup activated with helium gas from room temperature. The samples had mass of 3.67 mg, 0.67 mg and 0.92 mg with area $2.78 \times 10^{-6}$ m$^2$, $5.16 \times 10^{-7}$ m$^2$, and $6.97 \times 10^{-7}$ m$^2$ respectively. The experimental details, including sample preparation and orientation, are identical to those we reported previously [14], with the exception of the additional dc field used in this study.

![FIG. 1: Onset of superconductivity for different dc fields and at fixed pressure of 1.82 kbar, with overlaid fits. Legend indicates the applied field in Tesla for each curve.](image-url)
For four runs in the pressure cell at nominally zero pressure and zero field, we found an average $T_c = 2.29$ K. In each case the transition temperature measured in the pressure cell was within 24 mK of the value outside the pressure cell. In some cases the temperature shift can indicate the initial pressure applied during the experimental setup and cooling process, typically about 0.3 kbar [14, 15]. However, $T_c$ in CeCoIn$_5$ is not very sensitive to c-axis pressure, shifting by less than 17 mK/kbar [14, 16], so it gives little information on the pressure offset. Hence the values for applied pressure used in this paper do not include any offset for an initial pressure.

Figure 1 presents susceptibility curves near the onset of superconductivity for several applied fields, all at a pressure of 1.82 kbar. As indicated by the black lines in the figure, we determine $T_c$ by assuming the susceptibility is constant in the normal state and changes linearly just below $T_c$. As usual for superconductors, the onset $T_c$ shifts to lower temperature as the applied field increases. In addition, the transition decreases in magnitude and broadens in temperature. The magnitude change is due to flux lines threading the sample, which lead to incomplete diamagnetism. The broadening is governed by the temperature-dependence of the vortex pinning. As temperature falls and the vortices are more strongly pinned, the behavior becomes increasingly diamagnetic. We now turn to temperature sweeps below 400 mK and at fields above 4.5 Tesla. In this regime the susceptibility has not saturated but retains a temperature-dependence which decreases with increasing field.

We confirmed that changing the pressure causes no systematic shift in either $\chi_s$ or $\chi_n$. Nor does field shift $\chi_n$. Together these indicate that only $\chi_s$ is affected and only by tuning applied field. This corresponds to a decrease in superconducting transition magnitude with increasing field.

Susceptibility at much lower temperatures appears in Figure 2. The data shown were measured at 1.42 kbar uniaxial pressure. When we increase the dc field the superconducting susceptibility $\chi_s$ decreases in magnitude. We track this shift as a function of field by picking out $\chi$ at a fixed temperature from each of these data sets. Figure 3 shows some of the resulting $\chi$ vs $H$ curves. We repeat this procedure for each of the four temperatures indicated by the dashed lines of Figure 2. In each case we smooth the data by averaging $\chi$ in a 40 mK range centered at the desired temperature.

The upper frame of Figure 3 shows data at 300 mK for several different pressures. Each data set appears to have linear regimes, but also a kink where the slope changes abruptly. At each pressure we fit a function of exactly this form: two linear portions, with a kink at $(H_k, \chi_k)$ where the slope changes. We use four free parameters: $H_k$, $\chi_k$, and the slope on each side. We vary $H_k$ and $\chi_k$ manually, in steps of 0.01 Tesla and 0.00075, respectively. For each kink location, we solve for the slopes which best fit the data above and below $H_k$. We then select the kink location which minimizes the least squares error. The lines overlaid in the center plot are the result of this minimization. The lower plot indicates the location of $H_k$ and also the critical field $H_{c2}$ at 300 mK for each pressure. The two decrease in concert with increasing pressure, showing that most of the kink’s shift with pressure comes from the reduction of $H_{c2}$.

The main portion of Figure 4 shows $H_{c2}(T)$ at ambient pressure, with $H_k$ also plotted. As seen in the lower part of Figure 3 c-axis pressure reduces $H_{c2}$ at low temperatures. At 500 mK, the change in $H_{c2}(T = 0)$ is 0.04 T/kbar. Hydrostatic pressure has a somewhat larger effect, between 0.06 and 0.08 T/kbar [4, 17]. Interestingly, although increasing hydrostatic pressure also pushes the high-temperature, low-field portion of the $H_{c2}(T)$ dome up to higher $T_c$, this shift is almost absent with uniaxial pressure. We recently reported that the zero-field $T_c$ has little dependence on c-axis pressure, rising only about 20 mK to a maximum near 2 kbar, so it is not surprising that c-axis pressure also has little effect on the phase boundary at very low fields.

The inset of Figure 4 expands the high-field, low-temperature region. The solid curve is $H_{c2}(P = 0)$, normalized to 1 at $T = 0$. The normalized $H_{c2}$ curves at other pressures are nearly identical and are omitted from the graph. The symbols show the normalized kink field, $H_k(T, P)/H_{c2}(0, P)$. The kink agrees well with the proposed HFP boundary, which appears between 0.955 and 0.96 in other measurements [3, 6, 18, 19]. Several transitions of the vortex lattice have also been observed [18], but the highest of these is near 0.85, much lower than the
FIG. 3: Top: Susceptibility at 300 mK with fits overlaid. See text for details. All data except zero pressure are from the same sample and are shifted vertically for clarity. Bottom: Plot of the location of the kinks \( H_k \) obtained from the fits (filled circles), with \( H_{c2} \) at 300 mK also shown (open squares).

FIG. 4: Main graph presents measurements of \( H_{c2}(T) \) at zero pressure (solid circles) and a fit to these points. Shaded triangles indicated the kink locations \( H_k(T) \). Inset shows an expanded view in the region of \( H_k \) near the top of the dome. The curve \( H_{c2}(T) \) is normalized to 1 at \( T = 0 \), and \( H_k \) is shown for several applied pressures.

Under uniaxial pressure both \( H_{c2} \) and \( H_k \) decrease, by roughly the same factor. \( H_k \) is also approximately independent of temperature and forms a linear boundary well below \( H_{c2} \).

To complement the temperature sweeps described above we also performed field sweeps at fixed temperature and pressure. One might expect a kink as in Figure 3 to appear directly in this measurement. However, in fact we find no evidence of a kink. At all fields below \( H_{c2} \), the slope of \( \chi \) vs \( H \) in the field sweeps is comparable to its high-field value in the temperature-sweep data. The low-field slope in the temperature sweeps is notably larger. The sensitivity to the measurement history could explain why some of the previous experiments have found no sign of a transition to the HFP for c-axis field.

One possible source of the kink is influence from the AFM QCP, which at ambient pressure occurs at a field very close to \( H_{c2} \). However, under hydrostatic pressure the QCP field decreases about five times as fast as \( H_{c2} \), so that the QCP moves deep inside the superconducting phase. While its behavior has not been tracked under uniaxial pressure, there is no reason to believe that \( H_{c2} \) and \( H_{QCP} \) would remain close together. Since we find that \( H_k \) changes with uniaxial pressure only about 1.5 times as fast as \( H_{c2} \), the kink is probably not an indication of the QCP.

Another interpretation is in terms of thermally activated flux flow [20]. Our temperature sweeps are nearly field-cooled measurements. Although in practice we often do not exceed \( T_c \), we find no apparent difference in the low-temperature susceptibility between these temperature sweeps and those when the sample does begin in the normal state. The larger low-temperature slope for temperature sweeps indicates that the vortex pinning is stronger after field-cooling. A plausible explanation is that at higher temperatures the vortices fluctuate enough to find the most favorable pin sites, which they cannot do when the field changes at low temperature. In the HFP, there is no such annealing effect, and the pinning strength always has the smaller value.

A change in pinning strength could arise from antiferromagnetic order in the region above \( H_k \), which could interact with the superconductivity. More generally, any change in the order parameter can reasonably be expected to affect the vortex pinning. Several reports have been made along with theoretical descriptions suggesting this phase is the FFLO phase (see reference [19] for an overview). In the FFLO state periodic planar nodes appear perpendicular to the flux lines, leading to a segmentation of the vortices into pieces of length \( \Lambda = 2\pi/q \), where \( q \) is the quasiparticle wave vector [19] which factors into the order parameter \( \Delta \propto \sin q \cdot r \). The pieces are relatively free of each other and hence better able than conventional vortices to position themselves at pin centers. Hence pinning forces on the flux lines should increase in the FFLO phase. If the nodal planes reflect the crystal structure, then uniaxial pressure parallel to the flux lines would compress the segmentation length \( \Lambda \) and change the periodicity of the order parameter. Already strongly Pauli-limited, CeCoIn\(_5\) under uniaxial pressure becomes even more so than with hydrostatic pressure, possibly due to increased hybridization between the Ce-In layers. The way that \( H_k \) tracks \( H_{c2} \) suggests a possible connection with the electron spin population. Theoret-
tical models suggest redistribution of spin states in the vortex cores upon entering the FFLO state \[21\]. Namely, at the intersection of the nodal plane and the vortex core, excess spin states are emptied.

Ultrasound measurements of the HFP boundary \[22\] for in-plane field can also be interpreted in terms of vortex pinning. The vortices affect the ultrasound velocity more strongly in the HFP, which is consistent with increased pinning; one explanation is segmentation of the vortices into short, somewhat independent pieces that can better take advantage of low-density pin sites. Such segmentation could arise from the planar order parameter itself. The location and pressure dependence of the \(H_k\) boundary provide further evidence for the HFP phase for \(H \parallel c\) in CeCoIn\(_5\). An unanswered question is why the \(H_k\) boundary shows up with temperature sweeps and not field sweeps.

In conclusion, we have measured ac susceptibility response of CeCoIn\(_5\) in an applied field, as well as under uniaxial pressure. We find evidence for a phase boundary in agreement with previous indications of the transition to the HFP at zero pressure. We report a shift in this boundary with uniaxial pressure which roughly tracks \(H_{c2}\), decreasing in field at a rate about 50% faster than the decrease of \(H_{c2}\). For \(H_{c2}\) itself, pressure depresses the critical field at low temperatures but has little effect near \(T_c\). This differs from hydrostatic measurements but is consistent with the minimal effect of uniaxial pressure on \(T_c\) itself. The location and pressure dependence of the \(H_k\) boundary could support a wider range of pinning strengths that produces both effects.

In conclusion, we have measured ac susceptibility response of CeCoIn\(_5\) in an applied field, as well as under uniaxial pressure. We find evidence for a phase boundary in agreement with previous indications of the transition to the HFP at zero pressure. We report a shift in this boundary with uniaxial pressure which roughly tracks \(H_{c2}\), decreasing in field at a rate about 50% faster than the decrease of \(H_{c2}\). For \(H_{c2}\) itself, pressure depresses the critical field at low temperatures but has little effect near \(T_c\). This differs from hydrostatic measurements but is consistent with the minimal effect of uniaxial pressure on \(T_c\) itself. The location and pressure dependence of the \(H_k\) boundary provide further evidence for the HFP phase for \(H \parallel c\) in CeCoIn\(_5\). An unanswered question is why the \(H_k\) boundary shows up with temperature sweeps and not field sweeps.

In conclusion, we have measured ac susceptibility response of CeCoIn\(_5\) in an applied field, as well as under uniaxial pressure. We find evidence for a phase boundary in agreement with previous indications of the transition to the HFP at zero pressure. We report a shift in this boundary with uniaxial pressure which roughly tracks \(H_{c2}\), decreasing in field at a rate about 50% faster than the decrease of \(H_{c2}\). For \(H_{c2}\) itself, pressure depresses the critical field at low temperatures but has little effect near \(T_c\). This differs from hydrostatic measurements but is consistent with the minimal effect of uniaxial pressure on \(T_c\) itself. The location and pressure dependence of the \(H_k\) boundary provide further evidence for the HFP phase for \(H \parallel c\) in CeCoIn\(_5\). An unanswered question is why the \(H_k\) boundary shows up with temperature sweeps and not field sweeps.

### I. ACKNOWLEDGEMENT

This work was funded by the NSF through grant DMR-0454869.

---

[1] P.G. Pagliuso, et al., “Multiple phase transitions in Ce(Rh,Ir,Co)In\(_5\),” *Physica B*, 312-313, 129 (2002); [arXiv:cond-mat/0107266]

[2] F. Ronning et al., “Pressure study of quantum criticality in CeCoIn\(_5\),” *Phys. Rev. B* 73, 064519 (2006); [arXiv:cond-mat/0602089]

[3] T. Tayama et al., “Unconventional heavy-fermion superconductor CeCoIn\(_5\): dc magnetization study at temperatures down to 50 mK,” *Phys. Rev. B* 65, 180504R (2002).

[4] C.F. Miclea et al., “Pressure dependence of the Fulde-Ferrell-Larkin-Ovchinnikov state in CeCoIn\(_5\),” *Phys. Rev. Lett.* 96, 117001 (2006).

[5] M. Kenzelmann et al., “Coupled superconducting and magnetic order in CeCoIn\(_5\),” *Science* 321, 1652 (2008).

[6] B.-L. Young et al., “Microscopic evidence for field-induced magnetism in CeCoIn\(_5\),” *Phys. Rev. Lett.* 98, 036402 (2007); [arXiv:cond-mat/0608040]

[7] P. Fulde and R. A. Ferrell, “Superconductivity in a strong spin-exchange field,” *Phys. Rev.* 135, A550 (1964).

[8] A.I. Larkin and Yu.N. Ovchinnikov, “Inhomogeneous state of superconductors,” Zh. Eksp. Teor. Fiz. 47, 1136 (1964) [Sov. Phys. JETP 20, 762 (1965)].

[9] A. Bianchi et al., “First-order superconducting phase transition in CeCoIn\(_5\),” *Phys. Rev. Lett.* 89, 137002 (2002); [arXiv:cond-mat/0203310]

[10] H.A. Radovan et al., “Magnetic enhancement of superconductivity from electron spin domains,” *Nature* 425, 51 (2003); [arXiv:cond-mat/0304526]

[11] A. Bianchi et al., “Possible Fulde-Ferrell-Larkin-Ovchinnikov superconducting state in CeCoIn\(_5\),” *Phys. Rev. Lett.* 91, 187004 (2003); [arXiv:cond-mat/0304420]

[12] K. Kumagai et al., “Fulde-Ferrell-Larkin-Ovchinnikov state in a perpendicular field of quasi-two-dimensional CeCoIn\(_5\),” *Phys. Rev. Lett.* 97, 227002 (2006); [arXiv:cond-mat/0605394]

[13] X. Gratens et al., “Complex mixed state of the Pauli-limited superconductor CeCoIn\(_5\),” *Phys. Rev. B* 85, 054502 (2012); also see [arXiv:cond-mat/0608722]

[14] S.D. Johnson, R.J. Zieve, and J.C. Cooley, “Nonlinear effect of uniaxial pressure on superconductivity in CeCoIn\(_5\),” *Phys. Rev. B* 83, 144510, (2011); [arXiv:1010.4653]

[15] O.M. Dix et al., “Anisotropic dependence of superconductivity on uniaxial pressure in CeIn\(_5\),” *Phys. Rev. Lett.* 102, 197001 (2009); [arXiv:0908.4366]

[16] N. Oeschler et al., “Uniaxial pressure effects on CeIrIn\(_5\) and CeCoIn\(_5\) studied by low-temperature thermal expansion,” *Phys. Rev. Lett.* 91, 076402 (2003).

[17] T. Tayama et al., “Pressure dependence of the first-order superconducting phase transition in CeCoIn\(_5\),” *J. Phys. Soc. Jpn.* 74, 1115-1118 (2005).

[18] A.D. Bianchi et al., “Superconducting vortices in CeCoIn\(_5\): toward the Pauli-limiting field,” *Science* 319, 177 (2008).

[19] Y. Matsuda and H. Shimahara, “Fulde-Ferrell-Larkin-Ovchinnikov state in heavy fermion superconductors,” *J. Phys. Soc. Jpn.* 76, 051005 (2007); [arXiv:cond-mat/0702481]

[20] C.J. van der Beek, V.B. Geshkenbein, and V.M. Vinokur, “Linear and nonlinear ac response in the superconducting mixed state,” *Phys. Rev. B* 48, 3393 (1993).

[21] T. Mizushima, K. Machida, and M. Ichioka, “Topological structure of a vortex in the Fulde-Ferrell-Larkin-Ovchinnikov state,” *Phys. Rev. Lett.* 95, 117003 (2005); [arXiv:cond-mat/0504665]

[22] T. Watanabe et al., “High-field state of the flux-line lattice in the unconventional superconductor CeCoIn\(_5\),” *Phys. Rev. B* 70, 020506(R) (2004); [arXiv:cond-mat/0312062]