PHYSICAL REVIEW B **71**, 020503(R) (2005)

Evidence for the Fulde-Ferrell-Larkin-Ovchinnikov state in $CeCoIn_5$ from penetration depth measurements

C. Martin and C. C. Agosta*
Department of Physics, Clark University, Worcester, Massachusetts 01610, USA

S. W. Tozer, H. A. Radovan, E. C. Palm, and T. P. Murphy
The National High Magnetic Field Laboratory, 1800 East Paul Dirac Drive, Tallahassee, Florida 32310, USA

J. L. Sarrao
Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA
(Received 4 September 2003; revised manuscript received 27 September 2004; published 7 January 2005)

We report penetration depth and resistivity measurements on the heavy fermion superconductor $CeCoIn_5$ using a self-resonant tank circuit based on a tunnel diode oscillator. For magnetic fields applied near parallel to the *ab* planes and temperatures below 250 mK, two phase transitions were found. The lower field transition, within the superconducting state, is of a second order and we identify it as the transition from the ordinary vortex state to the Fulde, Ferrell, Larkin, Ovchinnikov (FFLO) state. The higher field transition marks the change from the FFLO to the normal state. This higher field transition, H_{c2}, is of first order up to 900 mK, the highest temperature measured. Our normal-state resistivity measurements at temperatures between 100 and 900 mK suggest that the FFLO state is related to the change of the quasiparticle interaction strength, F_0^a.

DOI: 10.1103/PhysRevB.71.020503 PACS number(s): 74.81.−g, 73.43.Fj, 74.25.Dw, 74.70.Tx

In 1964, Fulde and Ferrell[1] and Larkin and Ovchinnikov[2] predicted that in a purely Pauli limited superconductor, the magnetic field acting on the Cooper pair's spin can induce pairs with nonzero total momentum and, consequently, a spatially modulated order parameter. This so-called Fulde, Ferrell, Larkin, Ovchinnikov (FFLO) state can lead to an enhancement of the critical field up to 2.5 times the Pauli paramagnetic limit (H_P).[3] We have made penetration depth measurements suggesting that the FFLO state exists in the heavy fermion superconductor $CeCoIn_5$.

Orbital effects are reduced in heavy fermion materials[9,11] because their very low Fermi velocity (large effective mass) decreases the orbital magnetic-field coupling. Orbital effects can be further reduced in anisotropic superconductors by careful orientation of the applied magnetic field, although the FFLO state may exist even in the presence of weak orbital effects.[4] There is a growing interest in the theoretical study of this more realistic case,[5–10] and special interest in anisotropic quasi-two-dimensional (quasi-2D)[5,6,8–10] and *d*-wave superconductors,[5,8,10] such as $CeCoIn_5$. Unambiguous experimental evidence for the formation of the FFLO state has been reported only very recently[12] from specific heat and magnetization measurements on $CeCoIn_5$. More recently, a paper was published by Bianchi *et al.*, that supports this claim.[13] Most of the previous experimental results on $CeCoIn_5(T_c=2.3\text{ K})$[14] are in good agreement with the theoretical criteria for observing the FFLO state. It has been shown that the orbital pair breaking effect has to be small or completely absent, as measured by the Maki parameter $\alpha=\sqrt{2}(H_{c2}^0/H_P)$ (Ref. 15), where H_{c2}^0 is the orbital critical field,[16] to favor the FFLO state. This yields $\alpha \geqslant 1.8$ according to Ref. 4, although this calculation was partially based on the BCS theory and may not apply to non-*s*-wave superconductors. A number of groups have calculated α,[12,16] but the parameters for this ratio, particularly H_P, are difficult to measure. From magnetization studies of the critical field[17,18] it is clear that, when the magnetic field is applied parallel to the *ab* planes (the *ab* planes are perpendicular to the [001] direction) at low temperature, H_{c2} is a first-order transition, indicating that $CeCoIn_5$ is in the Pauli limit. We have calculated the Pauli limit for $CeCoIn_5$ by using a theory-independent method,[19,20] that requires use of Wilson's ratio.[21] The basis of the calculation is that the condensation energy, U_c, is related to both the specific heat via the specific-heat jump, and the binding energy of the Cooper pairs via

$$U_c = \frac{\mu_0}{2}\chi_e H_P^2, \tag{1}$$

where χ_e is the Pauli paramagnetic susceptibility. We propose that the jump in the measured specific heat can be integrated from the superconducting transition to zero temperature as an estimate of the condensation energy of the superconducting state, and, using the measured susceptibility, H_P can be calculated. Although the specific heat leads to a good measure of the condensation energy, the susceptibility measures the Pauli susceptibility plus unwanted terms such as the Landau diamagnetism and inner-core electrons. One way to isolate the Pauli susceptibility is to use the Sommerfeld constant γ to estimate χ_e through the use of Wilson's ratio. The difficulty with this method is that the Landé *g* factor[21] needs to be measured to find Wilson's ratio, and we are unaware of any direct measurements of *g* in $CeCoIn_5$. However, Won *et al.*[22] have recently published a critical-field calculation, which they fit to critical-field data with *g* as one of the few free parameters. In this paper they found $g=0.64$ in the parallel orientation and 1.5 in the perpendicular orientation. Using data for γ, ΔC, and $H_{c2}(0)$ ($H^0_{c2\parallel}=31.5$ T and $H^0_{c2\perp}=12.5$ T),[12,14,17] we calculated the Pauli limit in $CeCoIn_5$ to be 7.3 and 4.8 T, yielding $\alpha=6.1$ and 3.7 when B is parallel and perpendicular to the ab planes, respectively. Higher values of H_P can be justified if $CeCoIn_5$ is not a Fermi liquid as is discussed below. These Pauli limits show that with H perpendicular to the ab planes, where the critical field was found to be 5.0 T, H_{c2} is near the Pauli limit, but in the orientation with H along the ab planes H_{c2} is above the Pauli limit, consistent with an FFLO state.

The enhancement of the upper critical field in the FFLO state can be particularly substantial for a 2D superconductor[24] and de Haas-van Alphen data on $CeCoIn_5$ indicate a pronounced quasi-2D Fermi surface.[25,26] Specific-heat data,[23] and thermal-conductivity measurements,[27] suggest the presence of nodes in the superconducting gap, which is important to characterize the FFLO state.[3,9,11] Another requirement for the FFLO state is that the superconductor be in the *clean* limit, $l \gg \xi$, where l is the mean-free path of the quasiparticles and ξ is the superconducting coherence length. With $l \geqslant 810$ Å and $\xi \leqslant 58$ Å based on Refs. 23 and 17, $CeCoIn_5$ meets this requirement.

We present tunnel diode oscillator (TDO) measurements on $CeCoIn_5$ to fields of 18 T, at different orientations and temperatures between 60 and 900 mK. The TDO is a self-resonant tank circuit, where the sample is placed in the coil with the ab planes perpendicular to the ac magnetic field.[28] In this orientation, the penetration depth is measured parallel to the ab planes. Crystal platelets of $CeCoIn_5$ with approximate dimensions of 1.0-mm diam$\times$0.1-mm thick and 0.3-mm diam$\times$0.1-mm thick were placed in a 1.33-mm and a 0.35-mm diam coil, respectively. Details of the sample growth and characterization can be found in Ref. 14. A thermometer was placed on the rotating platform so that we were able to account for the dc power added by the TDO circuit. The data reported in this article comes from the large sample where the resonant frequency of the circuit at 80 mK was $\simeq 189$ MHz. The smaller sample and coil at 1.2 GHz yielded similar results. For a TDO experiment, the relative change in the resonant frequency is proportional to the change in the penetration depth $[\Delta f/f_0 = \eta(\Delta\lambda/\lambda_0)]$. Obtaining absolute values of the penetration requires a careful calculation of the constant η, which depends on the coil and sample geometries and the demagnetization factor. We did not calibrate the system for obtaining absolute values, but we measured and subtracted the influence of the background by running the system both with and without the sample. In the normal state, the penetration is limited by the skin depth ($\delta \propto \sqrt{\rho}$), and we were able to measure the change in resistivity with magnetic field and temperature.

Our H-T phase diagram for the field applied perpendicular to the ab planes is in very good agreement with the magnetization data[17] as is shown in Fig. 1. Analysis of our data shows that this transition changes from first to second order between 425 and 815 mK, where we no longer observed a sharp jump in the TDO frequency at H_{c2}, which is similar to other experiments.[16,17] To the lowest temperature measured, we do not see evidence for the FFLO state in the perpendicular direction, although we note that the shape of the 62.7-mK sweep is different than the higher temperature sweeps.

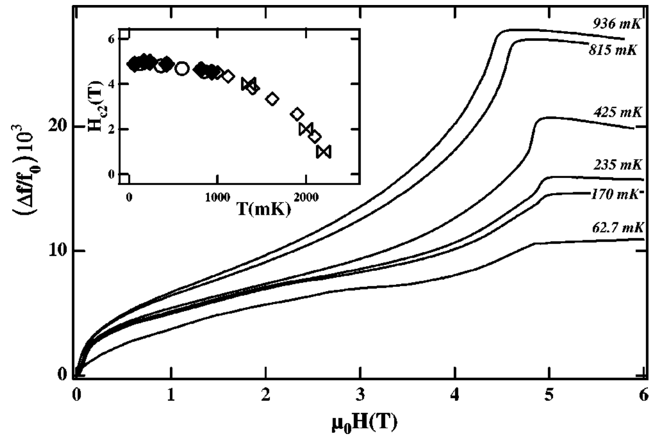

FIG. 1. The relative change in frequency with magnetic field at different temperatures for H perpendicular to the ab planes. The two sweeps above 800 mK do not feature a clear jump in frequency at H_{c2}. The inset displays the H_{c2} vs temperature for this orientation. The filled diamonds represent our data, and the open diamonds, circles, and bowties are magnetization data from Ref. 17.

Figure 2 shows the relative change in frequency with magnetic field when the field is applied parallel to the ab planes. In this orientation the contribution to the penetration depth due to the vortices is minimized in accordance with theory[29] and as is evidenced by the linear penetration low temperatures. As can be seen in Fig. 2, there is a lower field kink, or continuous transition reflected by the change of the slope of the penetration depth versus field, followed by a sharp jump at higher field. We assign the kink, a second-order transition, to the ordinary vortex state (VS)-FFLO transition and the upper transition, which is first order, to the FFLO-normal-state transition. Theoretically,[8,9,11] unlike originally predicted, the transition at the lower critical field could be of second order, as seen in our data. Above $T \simeq 250$ mK, the second-order transition is no longer present. The data shows only the first-order phase transition up to the

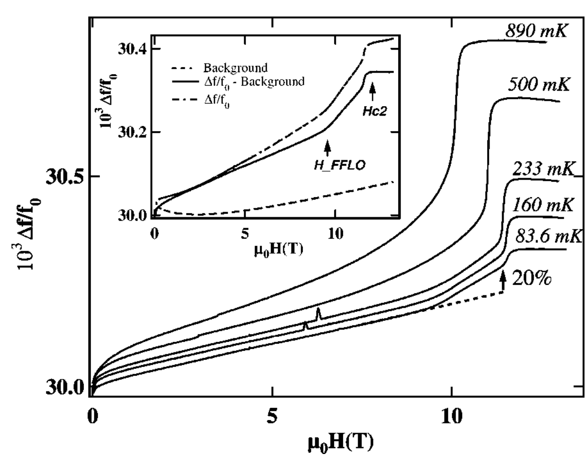

FIG. 2. The relative change in frequency with magnetic field at different temperatures for $H//ab$ planes. The dotted line in the main figure shows the trend set by the London penetration depth. The inset shows the raw data at 83.6 mK, and the influence of the empty coil background.

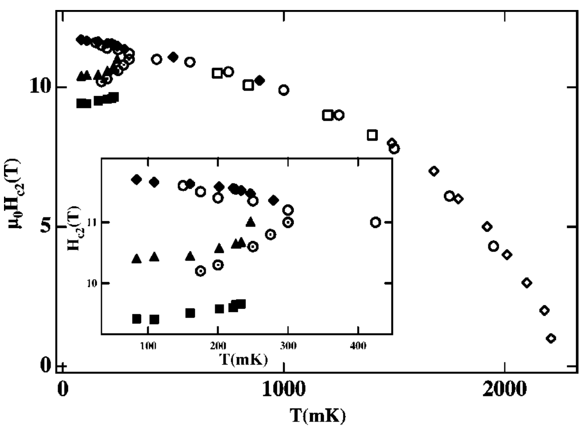

FIG. 3. Critical field as a function of temperature for $H//ab$. The filled symbols represent our data: the upper critical field (diamonds), the FFLO transition from the point of inflection (equilateral triangles), and the FFLO from the position of the *kink* (squares). Open symbols are data from other studies: specific heat from Ref. 12 (circles and dotted circles) and magnetization (Ref. 17) (diamonds) and (Ref. 18) (squares).

dilution refrigerator limit of 900 mK, as seen in Fig. 2. This first-order transition was predicted by Maki for a type-II superconductor with a large α parameter, below $0.56T_c$, albeit for the dirty limit.[15]

The distinguishing feature of the FFLO state is that the sign of the order parameter alternates spatially, either sinusoidally or more abruptly as the magnetic field approaches the FFLO-normal-state phase line.[11,30] The penetration depth, which is a function of the order parameter[31] ($\lambda \propto 1/|\psi|$ and $\propto 1/\sqrt{n_s}$), is sensitive to the density of superconducting electrons, n_s. If the order parameter oscillates and is no longer uniform, then the average density of superconducting electrons will be less and the penetration depth will increase. As a model we calculated the change in penetration if the order parameter started as a square wave at the VS-FFLO transition with the same amplitude as the VS state, to a sine wave at the FFLO-normal-state transition, based on Ref. 30. The result was that the penetration depth should increase by 20% beyond the trend that already exists in the VS. This is very close to what we measure, as shown in Fig. 2. It is important to mention that the VS-FFLO transition occurs within the superconducting state where resistance measurements, of course, cannot see any signal change.

In Ref. 12 the FFLO state has been observed up to $T \sim 350$ mK and B_{FFLO} is almost 1-T higher than the values we obtain. One possible explanation for the difference in the position of the VS-FFLO transition may be that the penetration depth and the specific heat measure different aspects of the VS-FFLO phase transition. To try and understand this discrepancy, we have calculated the second derivative of the penetration depth with respect to the field λ''. We see a peak in λ'' corresponding to the point of maximum curvature or the obvious kink in the penetration depth measurement near 9.5 T. The second easily identifiable feature in λ'' is where it crosses zero, signifying an inflection point. The zero of λ'' corresponds to the end of the second-order transition as measured by the TDO, and this point matches the transition as measured by specific-heat data (see Fig. 3).[12] Given that λ'' is monotonic and has no zero crossings when the angle is greater than 15° (see Fig. 4) or the temperature is greater than 250 mK, and that the change in the slope of the penetration depth is consistent with an oscillating order parameter, this data provides clear evidence of an FFLO state in $CeCoIn_5$.

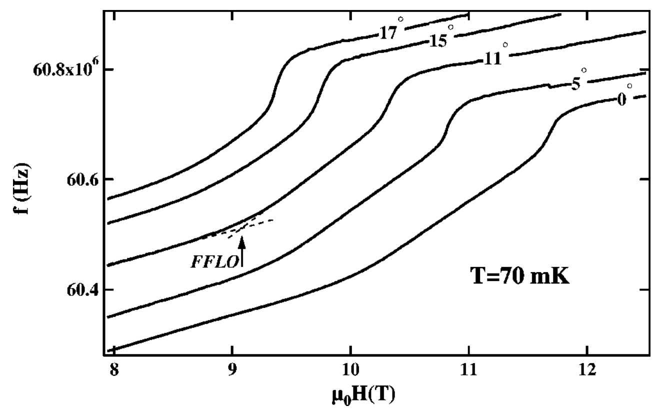

FIG. 4. The penetration depth as a function of magnetic field applied at different angles with respect to the *ab* planes. The FFLO transition goes away due to increasing orbital effects, however, the superconducting to normal-state transition remains first order, as also was found in Refs. 17 and 12.

As we have mentioned, the large jump in penetration depth that we associate with the first-order upper critical field matches very well with data previously obtained by other techniques, as seen in Fig. 3. Yet, as can be seen in Fig. 2, with increasing temperature, the height of the first-order transition increases. This increase is due to the change in resistivity of the normal state. By measuring the relative change in frequency with temperature, at a field of $B=12$ T parallel to the *ab* planes, we were able to observe the relative change in resistivity in the field-induced normal state (Fig. 5). For temperatures below $T \approx 300$ mK a variable power-law fit of the data yields a power of 1.92 which is close to 2, the value expected for a Fermi liquid (FL). In contrast, the two points at higher temperature depart from the power-law curve, indicating a change in the behavior of the system to a non-Fermi liquid (NFL). We are aware of very recent similar

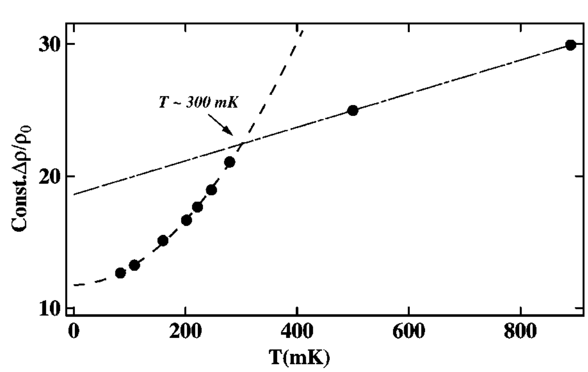

FIG. 5. Change in behavior of the normal-state resistivity with temperature at 12 T. The change suggests FL to NFL behavior as the temperature is increased. The lines are power law and linear fits.

results obtained from a direct measurement of the resistivity[32] and the Sommerfeld constant γ.[33] (Other penetration depth studies have also found evidence for NFL behavior, but at zero magnetic field[34,35]). At 5.5 T with the field perpendicular to the *ab* planes we only see a linear dependence of the resistivity indicating NFL behavior. This result is consistent with Refs. 32 and 33, where the crossover to FL behavior is observed above 7 T. This change in behavior leads us to believe that the same parameter may be responsible for the suppression of the FFLO state at higher temperatures *and* for the change in resistivity behavior. The coefficient F_0^a, which measures the interaction strength between quasiparticles, could be this parameter. A larger positive value of F_0^a, and therefore a stronger electron-electron interaction, results in a T^2 variation of resistivity with temperature,[36] and at the same time increases the range of stability of the FFLO state by lowering the Pauli paramagnetic susceptibility of the Fermi-liquid system ($\chi_e \simeq \chi_e 0/(1+F_0^a)$, where χ_e is the normal-fluid susceptiblility).[11] According to Burkhardt,[11] as F_0^a becomes smaller, the FFLO state is stable over a smaller temperature and field range and disappears for $F_0^a < -0.5$. It is interesting to note that $(1+F_0^a)^{-1} = R$, Wilson's ratio. Although the absolute value of R is unclear because of the problems of isolating the electron paramagnetic susceptibility, as mentioned above, below $T = 2$ K, χ_e is constant and γ increases and thus the trend is for R to decrease, which, consistent with Burkhart, stabilizes the FFLO state.

We would like to acknowledge R. McKenzie and N. Fortune for helpful discussions. Funding for this project came from the NHMFL In House Research and Visiting Scientist Programs. The NHMFL is supported by the NSF (DMR-0084173) and the State of Florida.

*Electronic address: cagosta@clarku.edu

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