Non- Locality and Strong Coupling in the Heavy Fermion Superconductor CeCoIn₅: A Penetration Depth Study

Elbert E. M. Chia, D. J. Van Harlingen, and M. B. Salamon

Department of Physics, University of Illinois at Urbana-Champaign, 1110 W. Green St., Urbana IL 61801

Brian D. Yanoff

General Electric, Schenectady, NY

I. Bonalde

Centro de Física, Instituto Venezolano de Investigaciones Científicas, Apartado 21827, Caracas 1020-A, Venezuela

J. L. Sarrao

Los Alamos National Laboratory, MST-10, Los Alamos, NM 87545

(Dated: November 3, 2018)

We report measurements of the magnetic penetration depth λ in single crystals of CeCoIn₅ down to ~0.14 K using a tunnel-diode based, self-inductive technique at 28 MHz. While the in-plane penetration depth tends to follow a power law, \( \lambda_{ab} / T^3 \), the data are better described as a crossover between linear (\( T \gg T^* \)) and quadratic (\( T \ll T^* \)) behavior, with \( T^* \) the crossover temperature in the strong-coupling limit. The c-axis penetration depth \( \lambda_c \) is linear in \( T \), providing evidence that CeCoIn₅ is a d-wave superconductor with line nodes along the c-axis. The different temperature dependences of \( \lambda_{ab} \) and \( \lambda_c \) rule out impurity effects as the source of \( T^* \).

The compounds CeMIn₅ (M = Co, Ir, Rh) have recently been added to the heavy-fermion family, and have attracted much interest due to their similarity with the cuprates: quasi-2D structure and proximity to magnetic order \( 1 \). CeCoIn₅, in particular, is a good candidate for study: its superconductivity is not sensitive to small changes in unit-cell volume or composition, unlike CeCu₂Si₂, and it has the highest \( T_c \) (~2.3 K) among the heavy-fermion superconductors. CeCoIn₅ has tetragonal HoCoGa₅ crystal structure, consisting of alternating layers of CeIn₃ and 'CoIn₂'. \( \lambda \) data revealed that the Fermi surface (FS) is quasi-2D, with an open 2D undulating cylinder extending along the [001] direction, as well as the large effective masses of electrons on this FS \( 2 \).

Recently, there has been mounting evidence for unconventional superconductivity in CeMIn₅. Specific heat data reveal a \( T^2 \) term at low temperature, consistent with the presence of line nodes in the superconducting energy gap \( 3 \). Thermal conductivity measurements with in-plane applied field show four-fold symmetry, consistent with nodes along the \( (\pm \pi, \pm \pi) \) positions \( 4 \). NQR measurements show that there is no Hebel-Slichter peak just below \( T_c \) \( 5 \). Below \( T_c \) the spin susceptibility is suppressed, indicating singlet pairing \( 6 \). However, there are some ambiguities in some of the measurements. Thermal conductivity data yield a \( T^3 \) low-temperature behavior, that the authors claim is close to \( T^3 \) behavior predicted for unconventional superconductors with line nodes in the clean limit \( 7 \). NQR measurements did not show the \( T^3 \) low-temperature behavior of \( 1/T \) that is expected for a line node gap; instead \( 1/T \) saturates below 0.3 K \( 8 \). Microwave measurements down to ~0.2 K showed a non-exponential behavior, and the authors claimed that \( \lambda(T) \sim T \) below 0.8 K \( 9 \), though the data clearly show some curvature in that temperature range. Further, the field was applied along the ab-plane, so the shielding currents have both in-plane and inter-plane components. In this paper, we present high-precision measurements of in-plane \( \lambda_{ab} \) and inter-plane \( \lambda_c \) penetration depths of CeCoIn₅ at temperatures down to 0.14 K. We find that \( \lambda_{ab} \) is best treated as a crossover from ~\( T \) to ~\( T^2 \) at a temperature \( T^* \). Combined with the result that \( \lambda_c \sim T \), this gives strong evidence for non-local behavior in a d-wave superconductor as predicted by Kostzin and Leggett \( 10 \).

Details of sample growth and characterization are described in Refs. \( 11, 12 \). Measurements were performed utilizing a 28 MHz tunnel diode oscillator \( 13 \) with a noise level of 1 part in 10⁸ and low drift. The magnitude of the ac field was estimated to be less than 5 mOe. The cryostat was surrounded by a bilayer Munetal shield that reduced the dc field to less than 1 mOe. The sample was aligned inside the probing coil in two directions: (1) \( ab \) plane perpendicular to the rf field, measuring the in-plane penetration depth \( \lambda_{ab} \) (screening currents in the \( ab \) plane); or (2) with the rf field parallel to the plane, giving a combination of \( \lambda_{ab} \) and \( \lambda_c \). The sample was mounted, using a small amount of GE varnish, on a rod made of nine thin 99.999% Ag wires embedded in Stycast 1266 epoxy. The other end of the rod was thermally connected to the mixing chamber of an Oxford Kelvinox 25 dilution refrigerator. The sample temperature is monitored using a calibrated RuO₂ resistor at low temperatures (\( T_{base} \sim 1.8 \) K), and a calibrated Cernox thermometer at higher temperatures (1.3 K - 2.5 K). We report data only for...
$T \geq 0.14$ K. The value of $T_c$ was determined from magnetization measurements to be 2.3 K, identical to the previously reported value [3].

The deviation $\Delta \lambda(T) = \lambda(T) - \lambda(0.14$ K) is proportional to the change in resonant frequency $\Delta f(T)$, with the proportionality factor $G$ dependent on sample and coil geometries. For a square sample of side $w$, thickness $2d$, demagnetization factor $N$, and volume $V$, $G$ is known to vary as $G \propto \frac{R_{3D}(1-N)}{V}$, where $R_{3D} = \frac{w}{2[1+(1+2d/w)^2]\arctan(w/2d)-2d/w}$ is the effective sample dimension [1]. For our sample $2w \approx 0.73$ mm and $2d \approx 0.09$ mm. We determine $G$ from a single-crystal sample of pure Al by fitting the Al data to extreme non-local expressions and then adjusting for relative sample dimensions. Testing this approach on a single crystal of Pb, we found good agreement with conventional BCS expressions.

Fig. 1 shows $\Delta \lambda_{ij}(T)$ as a function of temperature. We see that $\Delta \lambda_{ij}(T)$ varies strongly at low temperatures, consistent with the exponential behavior expected for isotropic $s$-wave superconductors. On the other hand, the variation is not linear, but has an obvious upward curvature, unlike the low-temperature behavior expected for pure $d$-wave superconductors. A fit of the low-temperature data to a variable power law, $\Delta \lambda_{ij}(T) = \alpha + cT^b$ yields $n = 1.43 \pm 0.01$ for sample 1 and $1.57 \pm 0.01$ for sample 2. The upper inset of Fig. 1 shows this approximate $T^{3/2}$ behavior for sample 1. Kosztin et al. have proposed a theory that gives a $T^{3/2}$ term from the gradual evolution of the pseudogap above $T_c$ to the superconducting gap below $T_c$. While resistivity measurements suggest the possibility of a pseudogap in CeCoIn$_5$ [5], which renders this interpretation feasible, a decrease in Knight shift was observed only starting at $T_c$ [6]. We take the latter to rule out a pseudogap mechanism.

Before considering novel excitation processes, we note the important distinction between $\Delta \lambda(T)$, which is directly measured, and the superfluid density $\rho(T) = \lambda^2(0)/\lambda^2(T)$, which can be inferred only with the knowledge of $\lambda(0)$ [10]. In the $d$-wave model, even if $\rho$ varies strictly with $T$, i.e. $\rho=1-\alpha T/T_c$, the penetration depth is non-linear: $\lambda(T) = \lambda(0)[1+1/2(\alpha T/T_c)+3/8(\alpha T/T_c)^2+...]$. Hence there is always a quadratic component to $\lambda$ whose strength depends on $\alpha$, which in the $d$-wave model, is inversely proportional to $d \Delta \langle \theta \rangle/\delta \theta |_{\text{node}}$, the angular slope of the energy gap at the nodes [11]. If $\rho(T)$ is linear in $T$, there is no need to invoke another mechanism.

To extract the in-plane superfluid density from our data, we need to know $\lambda_{ij}(0)$. For a quasi-2D superconductor with a cylindrical Fermi surface and the material parameters in Ref. [3][13], we obtain $\lambda_{ij}(0) = 2600$ Å, considerably larger than the experimentally obtained value of 1900 Å [7]. This along with a large heat-capacity jump at $T_c$ leads us to consider strong-coupling corrections as listed below [13, 20]:

$$\eta_{Cv}(\omega_0) = 1 + 1.8(\frac{\pi T_c}{\omega_0})^2(\ln(\frac{\omega_0}{T_c})+0.5);$$

$$\eta_{\Delta}(\omega_0) = 1 + 5.3(\frac{T_c}{\omega_0})^3\ln(\frac{\omega_0}{T_c});$$

$$\eta_{\lambda}(\omega_0) = \sqrt{1 + (\frac{\pi T_c}{\omega_0})^2(0.6\ln(\frac{\omega_0}{T_c}) - 0.26)}$$

$$\quad \frac{1 + (\frac{\pi T_c}{\omega_0})^2(1.1\ln(\frac{\omega_0}{T_c}) + 0.14)}{1 + (\frac{\pi T_c}{\omega_0})^2}$$

each $\eta$ represents the correction to the corresponding BCS value. If we take the experimental value of $\Delta C/T_c = 4.5$ [14], then Eq. 1 gives the characteristic (equivalent Einstein) frequency $\omega_0 = 9.1$ K and $\lambda_{ij}(0) = 1500$ Å. However, Petrovic et al. 8 argued that since $C/T$ increases with decreasing temperature, the specific heat coefficient $\gamma$ is temperature-dependent below $T_c$. This effect calls into question simple estimates of strong-coupling corrections for CeCoIn$_5$. A better estimate is to use $\Delta C/\Delta S$, where $\Delta S$ is the measured change in entropy of the sample from $T = 0$ to $T_c$. Ref. 8 then gives $\Delta C/\Delta S = 2.5$, so that $\omega_0 = 17.9$ K, resulting in $\Delta_0^c = 2.1k_B T_c$ and $\lambda_{ij}(0) = 2000$ Å. On the other hand, the larger $\Delta C$ of Ref. 21 yields $\Delta C/\Delta S = 2.8$ and $\omega_0 = 15.4$ K, leading to $\Delta_0^c = 2.2k_B T_c$ and $\lambda_{ij}(0) = 1900$ Å. These values of $\lambda_{ij}(0)$ are close to that obtained by Ormeno et al. 7.

Although we will argue that non-local effects are important, we will refer to $(\lambda_{ij}(0)/\lambda_{ij}(T))^2$ as the “superfluid density.” Fig. 2 shows the calculated behavior.
of that quantity using the three values of $\lambda_{//}(0)$ obtained above. We follow the procedure in Ref. 10 to compute the experimental superfluid density, using the $T^{0/2}$ fit to estimate the small difference between $\lambda_{//}(0)$ and $\lambda_{//}(0.14 \text{ K})$. In each case, $\rho(T)$ is clearly not linear in $T$.

Non-linearity in $\rho(T)$ can arise from a crossover from an intermediate-temperature (pure) linear-$T$ behavior to, for example, low-temperature (impurity-dominated) quadratic behavior as pointed out by Hirschfeld and Goldenfeld 22. They interpolated between these two regions using

$$\lambda = \lambda_0 + b T^2 / (T^* + T),$$

where $T^*$ is the crossover temperature. In terms of superfluid density, one obtains 10

$$\Delta \rho_{//}(T) = \frac{\alpha T^2 / T_c}{T^* + T},$$

where $T^*$ depends on impurity concentration.

A much more provocative source of the crossover of Eq. 4 was suggested by Kosztin and Leggett [KL] 8, who showed that for $d$-wave superconductors, nonlocal effects change the linear behavior to quadratic below a crossover temperature $T_{T_m\text{local}} = \Delta_0 \xi_{//}(0)/\lambda_{//}(0)$.

The solid lines in Fig. 2 are fits to Eq. 5 and are very good for all three values of $\lambda_{//}(0)$. The value of $\alpha$ varies from $0.5$ to $0.7$, the smallest value of $\alpha$ belonging to the largest value of $\lambda_{//}(0)$. The value of $\alpha$ obtained is similar to that found for YBa$_2$Cu$_3$O$_{6.95}$ (22) 24, but smaller than that of Tl$_2$Ba$_2$CuO$_{6+\delta}$ (22) 22 and K-(ET)$_2$Cu[N(CN)$_2$]Br (22) 14. The value of $T^*$ varies less across the three $\lambda_{//}(0)$ values, from $0.32 \text{ K}$ to $0.42 \text{ K}$.

These values of $T^* / T_c$ (~0.14 - 0.18) differ from the cuprates 25, 25, 26 and the organic superconductor K-(ET)$_2$Cu[N(CN)$_2$]Br (~0.05), where impurity scattering is presumed to be the source. Further, Ref. 3 puts an upper limit of $20 \text{ ppm}$ on the impurity concentration. In the dirty $d$-wave model 24, this gives the unitary-limit scattering rate $\Gamma \sim 1.5 \times 10^8 \text{ s}^{-1}$, which yields an upper limit for $T^* \sim 65 \text{ mK}$. This is about 5 times smaller than the experimentally obtained values above, suggesting that the sample is too clean for the dirty $d$-wave model to be applicable.

Having ruled out impurity scattering, we turn to nonlocal electrodynamics as the source of the crossover in $\rho_{//}(T)$. For a $d$-wave superconductor with line nodes along the $c$-axis, nonlocality is expected to be relevant only when the applied magnetic field is oriented parallel to the $c$-axis, while the effect of impurities should not depend on the orientation of the field. As KL noted, if $T^*$ is noticeably smaller for $H \perp c$ than for $H_{//c}$ we may conclude that the observed effect is due mainly to nonlocal electrodynamics and not to impurities. For $H \perp c$, screening currents flow both parallel and perpendicular to the $c$-axis, mixing $\lambda_{//}$ and $\lambda_{\perp}$ with the frequency shift given by $\Delta f_{\perp} = \frac{V_0}{2V_f} \left( \frac{\lambda_{//}}{d} + \frac{\lambda_{\perp}}{d} \right)$ 11, where $V_0$ is the effective coil volume and $f_0$ the resonant frequency with the sample absent. In order to extract $\lambda_{\perp}$ we subtract out the $\lambda_{//}$ component from $\Delta f_{\perp}$. Fig. 3 shows the inter-plane penetration depth $\lambda_{\perp}$ of CeCoIn$_5$ down to $0.14 \text{ K}$. It is clearly linear in $T$ from $0.14 \text{ K}$ to $1 \text{ K}$. To obtain the superfluid density, we estimate $\lambda_{//}(0)$ from the $H_{c2}$ anisotropy of $\sim 2.3$ 21, and the fact that $\lambda(0) \propto \sqrt{H_{c2}(0)}$ 27, obtaining $\lambda_{//}(0) \sim 2700 \text{ Å}$. This is close to the value of $\sim 2800 \text{ Å}$ obtained from microwave mea-
measurements in the planar geometry \cite{28}. If we fit $\lambda_\parallel(T)$ to Eq. \cite{14}, we find $T_\parallel^* \lesssim 0.15$ K, significantly smaller than 0.32 K obtained for the in-plane case. This satisfies the Kosztin-Leggett test and indicates that the superfluid response of CeCoIn$_5$ is governed by nonlocal electrodynamics. This is also strong evidence that CeCoIn$_5$ is a $d$-wave superconductor with line nodes along the $c$-axis. Sr$_2$RuO$_4$ failed this test \cite{10} because its line nodes are horizontal instead of vertical. Kusunose and Sigrist argued that horizontal line nodes give power-law behaviors with less angular dependence for any inplane direction of the screening currents, and hence applied field \cite{10}. A calculation of $\rho_\perp$ is shown in the inset of Fig. \ref{fig:2}, the upturn below 0.5 K is an artifact of the choice of $\lambda_c(0)$. A larger value of $\lambda_c(0)$ would remove this feature, but there is no justification for doing so.

As a final test of the nonlocal scenario, we estimate $T_\perp$ using strong-coupling parameters. From the measured $H_{c2}(0)[001]$ value of 49.5 kOe, the coherence length $\xi_{\parallel}(0)$ is calculated to be 82 Å \cite{24}. Together with the earlier-derived values of $\Delta_0^c = 2.2k_BT_c$ and $\lambda_{\parallel}^c(0) = 1900$ Å, we find the strong-coupling nonlocal crossover temperature $T_{\text{nonlocal}}^* = \Delta_0^c / \lambda_{\parallel}^c(0) = 0.22$ K. Using a weak-coupling $d$-wave $\Delta(0) = 2.14k_BT_c$, we find $T_{\text{nonlocal}} = 0.26$ K. We regard either value to be satisfactorily close to the experimental value of 0.32 K. Note that the value of $\xi_{\parallel}(0)$ is different from the calculated BCS value of 58 Å \cite{24} or the strong-coupling corrected value of $\sim 50$ Å \cite{19}. This is not surprising since the BCS expressions \cite{19} assume a spherical FS, while LDA band structure reveals a very complicated FS with contributions from three different bands \cite{30}.

In conclusion, we report measurements of the magnetic penetration depth $\lambda$ in single crystals of CeCoIn$_5$ down to $\sim 0.14$ K using a tunnel-diode based, self-inductive technique at 28 MHz. The in-plane penetration depth ($\lambda_{\parallel}$) exhibits a crossover between linear (at high $T$) and quadratic (low $T$) behavior with a crossover temperature $T_{\text{nonlocal}}^* \approx 0.32$ K. Such behavior can arise in a superconductor with nodes in the gap either in a dirty $d$-wave model or from nonlocal electrodynamics. The linear low-temperature dependence of the $c$-axis penetration depth $\lambda_\perp$ strongly favors the nonlocal model with line nodes parallel to the $c$-axis. We also demonstrate that strong-coupling corrections are required to reconcile various experimentally determined superconducting parameters.

One of the authors (E.E.M.C.) acknowledges R. Prozorov and Q. Chen for useful discussions. This work was supported by the NSF through Grant No. DMR9972087. Work at Los Alamos was performed under the auspices of the U.S. Department of Energy.

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