CeCoIn$_5$ — a quantum critical superfluid?

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We have made the first complete measurements of the London penetration depth $\lambda(T)$ of CeCoIn$_5$, a quantum-critical metal where superconductivity arises from a non-Fermi-liquid normal state. Using a novel tunnel diode oscillator designed to avoid spurious contributions to $\lambda(T)$, we have established the existence of intrinsic and anomalous power-law behaviour at low temperature. A systematic analysis raises the possibility that the unusual observations are due to an extension of quantum criticality into the superconducting state.

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A number of experiments on the recently discovered family of heavy fermion superconductors CeRhIn$_5$, CeIrIn$_5$ and CeCoIn$_5$ point to the existence of a non-Fermi-liquid (NFL) metallic state in these compounds. Central to this conclusion is the observation, in CeIrIn$_5$ and CeCoIn$_5$, that two of the key parameters of a Fermi liquid — the electronic heat capacity coefficient $\gamma = C/T$ and Pauli susceptibility $\chi$ — increase on cooling and show no sign of entering a temperature-independent Fermi-liquid regime. Similar behaviour is observed in other heavy-fermion materials and is understood in terms of magnetic fluctuations near a zero-temperature critical point, where theory predicts either $\gamma \sim -\ln T$ or $\gamma = \gamma_0 - AT^{1/2}$, depending on the dimensionality and nature of the magnetism. What happens to such a system when it becomes superconducting is at present an open question. Do the Fermi liquid parameters continue to evolve within the superconducting state, or does superconductivity abort the approach to quantum criticality? Also, would the absence of quantum-critical superfluidity in materials such as the cuprate superconductors rule out a zero-temperature critical point as the source of magnetic fluctuations near a normal-state behaviour? These are important issues, which we address in this Letter with the first complete and well controlled measurements of the London penetration depth $\lambda(T)$ of a superconductor known to be situated near a magnetic quantum critical point. Our novel oscillator design and carefully characterised samples avoid extrinsic contributions to the $\lambda(T)$ signal, and a systematic analysis leads us to consider new, NFL physics as the reason for the anomalous power-law behaviour observed in $\lambda(T)$ in the low temperature limit.

The normal phase of CeCoIn$_5$ contains all the hallmarks of quantum criticality. De Haas-van Alphen (dHvA) measurements reveal large cyclotron masses that are strongly field dependent, exceeding 100 $m_0$ at low fields. The resistivity is reminiscent of the cuprates, nearly linear in temperature below 20 K. In magnetic fields large enough to suppress superconductivity, $C/T \sim -\ln T$ down to the lowest temperatures. From 2.5 K up to 100 K the spin–lattice relaxation rate has the $T^{1/4}$ temperature dependence expected near an antiferromagnetic instability, indicating proximity to a zero-temperature magnetic critical point. This is supported by the fact that CeCoIn$_5$ is a higher-density analogue of CeRhIn$_5$, a material in which weak, ambient-pressure antiferromagnetism gives way to non-s-wave superconductivity with $T_c = 2.1$ K at a pressure of 1.6 GPa.

The superconducting state of CeCoIn$_5$ is also highly unconventional and appears to bear strong similarities to that of the cuprate superconductors. Measurements of specific heat and thermal conductivity $\kappa(T)$ reveal low temperature power-law behaviour, similar to expectations for a pairing state with line nodes. A strong-power law temperature dependence is also observed in the low-temperature microwave surface impedance. The four-fold modulation of $\kappa$ by an angle-dependent basal-plane magnetic field indicates that the line nodes lie along the [110] directions. This is confirmed by a field-rotation study of dHvA oscillations, which increase in amplitude below $H_{c2}$ for fields in the [110] direction. In addition, observations of Pauli-limited superconductivity in $\kappa(H)$ and a Knight shift that decreases below $T_c$ in all directions imply spin-singlet superconductivity. Together the experiments strongly suggest that pairing occurs in a $d_{x^2−y^2}$ state.

CeCoIn$_5$ forms in a tetragonal crystal structure with alternating layers of CeIn$_3$ and CoIn$_2$, and superconducts at ambient pressure below $T_c = 2.25$ K. The high-quality CeCoIn$_5$ crystals used in our experiment were grown by a self-flux method in excess In. Although the start-
ing materials are very pure to begin with (Ce: 99.99%, Co: 99.9975% and In: 99.9995%), growth in excess In is expected to further refine the crystals. These naturally form as large ab-plane platelets, with mirror-like surfaces, and are ideally suited to high-frequency measurements. The high homogeneity and low defect level of the crystals are confirmed by our microwave measurements, which show a sharp superconducting transition ($\Delta T_c < 30$ mK) and a low quasiparticle scattering rate ($1/\tau$, the width of the conductivity spectrum $\sigma(\omega)$, $\approx 2 \times 10^{10}$ s$^{-1}$ at 1.2 K)

We focus on data from one sample, a crystal with dimensions $a \times b \times c = 1.38 \times 1.37 \times 0.073$ mm$^3$.

The $\lambda(T)$ measurements reported here were made with a 130 MHz tunnel diode oscillator (TDO) operated in $^3$He and dilution refrigerator cryostats over the temperature range 0.1 K to 9 K. In addition, a thorough study of the microwave conductivity $\sigma(\omega,T)$, obtained from surface impedance measurements from 1 to 75 GHz, was made on the same crystals used in the TDO experiments. Although the microwave measurements form an important part of this work (as a sample characterisation tool, for cross-checking and calibrating the TDO measurements, and as a means of determining the absolute penetration depth) a detailed discussion of those results will be presented elsewhere [4]. We focus here instead on describing the distinctive features of our tunnel diode oscillator and the results obtained using it.

A good description of the general principles of operation of the TDO is given in Ref. [2], and oscillators of this sort have been used by other workers to study unconventional superconductors [4, 7]. The chief innovation in our apparatus is the novel geometry of the oscillator’s probe circuit — a very small, self-resonant superconducting coil that acts as a local probe of the magnetic penetration depth. The miniature resonator has a square cross-section of side 0.5 mm and is wound from superconducting Nb wire on a high purity sapphire former. The high quality factor of the coil ($Q = 2.5 \times 10^5$ at 1.3 K) allows the resonator to be coupled inductively to the tunnel diode, with the mutual inductance backed-off until the diode only marginally sustains oscillation. This results in a frequency stability better than 1 part in $10^9$ per hour and a field at the sample of $\sim 10$ nT.

The combination of a high-$Q$ resonator and low-power tunnel diode keeps the total heat load of the oscillator below 2 $\mu$W and allows the whole circuit to be cooled into the mK temperature range. In the experiments, a basal-plane face of the single-crystal sample is attached to one end of the sapphire former with vacuum grease, about 50 $\mu$m from the end of the coil. In this geometry, the coil locally induces currents that flow in a 0.5 mm wide ring in the centre of the crystal face, as shown in the inset of Fig. 1. This is important when studying the superfluid response of electrically anisotropic materials such as CeCoIn$_5$, for two reasons: first, only basal-plane currents flow, eliminating contamination of the signal by c-axis currents; secondly, this experimental geometry is inherently insensitive to nonlocal effects, as electrons in a quasi-2D metal are intrinsically confined to move parallel to the basal plane [13].

In our setup there is thermal contact between the sample and the resonator, with the result that the temperature of the entire oscillator is swept during the experiments. Being able to operate in this mode has the potential advantage that both resonator and sample can be embedded in a hydrostatic pressure medium, in principle allowing the investigation of other quantum-critical systems, such as CePd$_2$Si$_2$ and CeIn$_3$ [19], which only superconduct under high pressures. However, changing the temperature of the oscillator also introduces systematic errors into the frequency shift signal. Fortunately, these are small over most of the temperature range and are highly reproducible, allowing them to be accurately accounted for using background measurements made in the absence of the sample. $\Delta \lambda(T)$ is obtained from the oscillator frequency-shift signal $\Delta f_0(T) = f_0(T) - f_0(T_{base})$ using the cavity perturbation approximation, where $\Delta \lambda(T) = -\Gamma \Delta f_0(T)$. Here $\Gamma = 6$ $\Lambda$/Hz is a temperature-independent geometric factor determined empirically by comparison with penetration depth measurements made down to 1.2 K using a 1 GHz loop-gaps resonator [20].

The absolute value of $\lambda(T)$ has been determined from measurements of the surface impedance $Z_s = R_s + iX_s$, made at 5.5 GHz with a TE$_{011}$ mode dielectric resonator. It is usually very difficult to determine $\lambda$ in this way, because only shifts in $X_s$ with temperature are experimentally accessible. However, by carrying measurements of $\Delta X_s(T)$ up to temperatures where the electronic scat-
terating rate is much larger than the microwave frequency (i.e. for $T \geq 20$ K in CeCoIn$_5$) we access the Hagen–Rubens limit where $R_s \approx X_s$. This enables the unknown offset in $X_s$ to be determined and, for sufficiently accurate $\Delta X_s(T)$ data, gives $X_s(T)$ absolutely down to 1.2 K. (Confidence in our procedure is enhanced by the fact that $R_s$ and $X_s$ match between 20 K and 90 K, and that an analysis of the departure of $R_s$ from $X_s$ below 20 K reveals a temperature-dependence of the optical effective mass that follows the ln $T$ behaviour of $C/T$ [2].) Having the $X_s(T)$ data is not enough: only in the low frequency limit, $\omega \tau \ll 1$, does $X_s = \omega \mu_0 \lambda$; at higher frequencies thermally excited quasiparticles also contribute to $X_s$ [2]. We use bolometric measurements of $\sigma(\omega, T)$ [22] in the superconducting state to properly account for the quasiparticle contribution, obtaining $\lambda(1.2$ K) = 3610 Å and $\lambda_0 = 2810$ Å in the $T \to 0$ limit. This is a large penetration depth, characteristic of a metal with heavily renormalised electrons.

Figure 3 shows the absolute $\lambda(T)$ data measured with the tunnel diode oscillator down to 0.1 K. In Fig. 3, a close-up view of the low-$T$ data reveals a strong temperature dependence, indicating low-lying excitations and supporting the case for line nodes in the pairing state. However, $\lambda(T)$ does not have the simple linear $T$ dependence expected for a $d$-wave superconductor and observed in the cuprates [20]; instead the inset to Fig. 3 reveals that $\lambda(T)$ is better approximated by a $T^{1.5}$ power law, down to 0.1 K. (Similar curvature in $\lambda(T)$ has been inferred from 10 GHz $X_s(T)$ measurements and attributed to disorder [12]. In those measurements, made down to only 0.25 K, $\omega \tau \approx 5$ at low $T$ and the clear connection between $X_s$ and the London penetration depth is lost.)

Also relevant is the observation that $\kappa(T) \sim T^{3.37}$ below 0.2 K [1]. The expected power law in the case of dilute strong-scattering impurities is $\kappa(T) \sim T^3$ [23], and an interesting possibility is that the fractional power laws in $\kappa(T)$ and $\lambda(T)$ might have the same origin. In Fig. 3, the data are plotted as $\rho_s(T) = \lambda^2(0)/\lambda^2(T)$. As $\rho_s$ is proportional to the ratio of the superfluid density $n_s$ to the effective mass $m^*$, it is expected to have a linear $T$ dependence in a $d$-wave superconductor, a geometric consequence of the presence of line nodes in the energy gap. This is not seen in our data, which show curvature over the full temperature range.

We now consider possible explanations for the observed $T$-dependence of $\lambda$, beginning with disorder. Strong-scattering impurities in a $d$-wave superconductor are known to induce a crossover in $\lambda(T)$ from clean-limit, $T$-linear behaviour above $T^* \approx 0.56\sqrt{n_1 T_F}$ to a quadratic $T$ dependence at low temperatures [24]. (Here $n_1$ is the density of impurities and $T_F$ the Fermi temperature.) We have assessed this possibility by fitting the interpolation formula of Ref. [24] $\Delta \lambda(T) \propto T^2/(T + T^*)$, to our data in Fig. 3 and obtain $T^* = 0.3$ K. Using $T_F = 50$ K (from specific heat [1]) we infer from $T^*$ that $n_1 = 0.26\%$. This is an order of magnitude greater than the density of impurities in our starting materials. Thermal conductivity experiments suggest the level of strong-scattering defects in our crystals is actually much lower: in $\kappa(T)$, $T^* < 30$ mK, implying $n_1 < 26$ ppm [11].

Alternatively, Kosztin and Leggett [18] have pointed out that nonlocal effects in a $d$-wave superconductor can cause an intrinsic crossover to $T^2$ behaviour in $\lambda(T)$, at
\( T^* \approx 2T_c \xi_0/\lambda_0 \), where \( \xi_0 \) is the BCS coherence length. Physically this is because the spatial extent of the Cooper pair is larger than \( \lambda_0 \) for a small range of angles around the nodal directions, and affects the field screening at correspondingly low temperatures. An estimate using the published value \( \xi_0 = 82 \text{ Å} \)\(^{[2]}\), gives \( T^* \approx 130 \text{ mK} \), significantly lower than the value obtained from the fit to our data in Fig. 2. In addition, for a quasi-2D \( d_{x^2−y^2} \) superconductor such as CeCoIn\(_5\), nonlocal effects should be very sensitive to geometry, being strongest for [100] surfaces, vanishing on [110] surfaces, and becoming extremely small for [001] surfaces, where the 2D electronic structure forces the electrons to move almost parallel to the crystal face. Our penetration depth probe takes advantage of this fact, only inducing currents on a [001] surface. In that case, \( T^* \) is determined by the out-of-plane coherence length, \( \xi \approx 35 \text{ Å} \)\(^{[5]}\), with the result that nonlocality should only become a consideration in our geometry below 56 mK.

In a multi-band metal there is the possibility that superconductivity occurs strongly for only some pieces of the Fermi surface, and is induced in the others by an internal proximity effect. This is thought to be relevant to CuO-chain superconductivity in the cuprates\(^{[23, 24]}\) and to the physics of Sr\(_2\)RuO\(_4\)\(^{[27]}\). In all cases it is expected to lead to a stronger \( T \)-dependence of \( \lambda(T) \), due to the presence of a small temperature scale associated with intrinsically weak superconductivity in parts of the Fermi surface. For CeCoIn\(_5\), which appears to be a \( d_{x^2−y^2} \) superconductor with line nodes, a proximity effect should result in positive curvature of \( \rho_s(T) \)\(^{[25]}\), opposite to that observed.

The Bose–Einstein condensation theory of Ref.\(^{[28]}\) predicts \( \Delta \lambda(T) \sim T^{1.5} \) at low \( T \), and has been applied to \( \lambda(T) \) measurements on the organic superconductor BEDT(TTF)\(_2\)CuC\(_5\)N\(_4\)Br\(_2\)\(^{[1]}\). The theory has been developed for the case of nonoverlapping Cooper pairs in a short-coherence-length, \( s \)-wave superconductor, as a possible explanation of the physics of the underdoped cuprates. It is not likely to be relevant to the case of CeCoIn\(_5\), which has strongly overlapping, \( d \)-wave pairs.

As none of the scenarios presented so far seems to provide an adequate account of the \( \lambda(T) \) data we propose another possibility, motivated by the NFL properties of the normal state. There we know that \( \gamma, \chi \) and \( m^* \) continue to be renormalised down to the lowest temperatures. If a \( T \)-dependent renormalisation also took place within the superconducting state, what would its effect on \( \rho_s(T) \) be? This question is complicated by the fact that the different bands in CeCoIn\(_5\) have (field-dependent) cyclotron masses spanning the range 10 to 100 \( m_0 \)\(^{[5]}\), and that \( \rho_s \propto (1/m^*) \). Nevertheless, an effective mass that increased with decreasing temperature would introduce the right curvature into \( \rho_s(T) \). To illustrate this point better we have taken our \( \rho_s(T) \) data, assumed the \( d \)-wave form for the superfluid density, \( n_s(T) = n_0(1−\alpha T/T_c) \), and plotted, below 0.8 K, in the inset of Fig. 3, the temperature dependence of \( m^* \) for different values of \( \alpha \). In each case \( m^*(T) \) shows a cusp-like upturn at low \( T \), reminiscent of the quantum-critical behaviour of \( C/T \) in the normal state. Are there other data to support this conjecture? A \( T \)-dependent renormalisation in the superconducting state should show up most clearly in the specific heat. In CeCoIn\(_5\) the measurement is complicated by the large low-\( T \) entropy of the In nuclei but, when the nuclear contribution is subtracted from the data, \( C/T \) still deviates from the expected \( d \)-wave \( T \)-linear behaviour, even showing signs of a low temperature upturn\(^{[4, 5]}\). The unusual low-\( T \) power law in the thermal conductivity, \( \kappa(T) \sim T^{3.37} \), might also find a consistent explanation within this scenario. (As with \( \rho_s(T) \), \( \kappa(T) \) would be dominated by the least divergent band.) Further measurements of thermodynamic properties in the low-\( T \) limit should help assess the validity of this proposal.

In conclusion, we have presented the first complete measurements of the London penetration depth of CeCoIn\(_5\) in the low-\( T \) limit and observed strong power-law behaviour that confirms the presence of low-lying excitations but is inconsistent with standard models of \( d \)-wave superconductivity. By working with carefully characterised samples and using a novel resonant-circuit probe that is inherently insensitive to nonlocal effects and electronic anisotropy we have been able to rule out extrinsic contributions to the \( \lambda(T) \) signal. We propose an alternative interpretation, and suggest that the NFL renormalisation occurring in the normal state of CeCoIn\(_5\) might also take place within the superconducting phase, leaving us with the exciting possibility that CeCoIn\(_5\) may be the first example of a quantum-critical superfluid.

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More recent measurements from E. Chia et al., which have come to our attention while preparing this work, confirm \( \Delta \lambda(T) \sim T^{1.5} \) at low \( T \), using crystals from the same source as ours.

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