Evidence for a Nodeless Gap from the Superfluid Density of Optimally Doped \( \text{Pr}_{1.855}\text{Ce}_{0.145}\text{CuO}_{4-y} \) Films

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We present measurements of the \( ab \)-plane magnetic penetration depth, \( \lambda(T) \), in five optimally doped \( \text{Pr}_{1.855}\text{Ce}_{0.145}\text{CuO}_{4-y} \) films for \( 1.6 \text{ K} \leq T \leq T_c \sim 24 \text{ K} \). Low resistivities, high superfluid density values \( n_s(T) \propto \lambda^{-2}(T) \), high \( T_c \)'s, and small transition widths are reproducible and indicative of excellent film quality. For all five films, \( \lambda^{-2}(T)/\lambda^{-2}(0) \) at low \( T \) is well fitted by an exponential temperature dependence with a gap, \( \Delta_{\text{min}} \), of 0.85 \( k_B T_c \). This behavior is consistent with a nodeless gap and is incompatible with \( d \)-wave superconductivity.

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It is widely accepted that pairing symmetry in the hole-doped cuprates is predominantly \( d_{x^2-y^2} \), at least near optimal doping where most phase-sensitive measurements have been made. Essentially all other experimental results agree with this view. The developing situation in the electron-doped cuprates is not so clear. While recent phase-sensitive measurements on optimally doped \( \text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y} \) (NCCO) and \( \text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y} \) (PCCO) films are consistent with a \( d_{x^2-y^2} \) energy gap, there is substantial evidence for a nodeless gap. The penetration depth, \( \lambda(T) \), measured by Aff et al. \(^{[6]} \) via field modulation of Josephson junctions fabricated in the \( ab \)-plane of optimally doped PCCO films is exponentially flat at low \( T \), suggesting a gapped superconducting state. The zero-bias peak seen in tunnelling data from hole-doped cuprates – associated with Andreev bound states of a \( d \)-wave order parameter – is absent from tunnel junctions in NCCO \(^{[11]} \), suggesting \( s \)-wave superconductivity. In light of perspicuous evidence for \( d_{x^2-y^2} \) superconductivity from phase-sensitive \(^{[6]} \) and angle resolved photoemission spectroscopy \(^{[7]} \) measurements on electron-doped samples, one might suspect some problem with measurements on in-plane tunnel junctions, or with the films themselves. This suspicion may be strengthened by reports of quadratic behavior of \( \lambda(T) - \lambda(0) \) at low \( T \) in optimally doped PCCO crystals \(^{[11]} \). Thus the importance of penetration depth measurements that confirm the findings of Aff et al. and demonstrate high sample quality and sample-to-sample reproducibility.

We present measurements of the \( ab \)-plane superfluid density, \( n_s(T) \propto \lambda^{-2}(T) \), in five optimally doped \( \text{Pr}_{1.855}\text{Ce}_{0.145}\text{CuO}_{4-y} \) films. \( \lambda^{-2}(T) \) at low \( T \) in hole-doped cuprates is consistently linear \(^{[10]} \) \(^{[11]} \) or quadratic \(^{[12]} \) in temperature. Theory has not found a scenario in which behavior flatter than \( T^2 \) is predicted for \( d_{x^2-y^2} \) superconductors. On the other hand, \( \lambda^{-2}(T) \) in gapped – e.g., \( s \)-wave – superconductors is exponentially flat at low temperatures. As shown below, \( \lambda^{-2}(T) \) in our optimally doped PCCO films is exponential in \( \Delta_{\text{min}}/T \), with the same value of the minimum gap, \( \Delta_{\text{min}} = 0.85 \pm 0.05 \, k_B T_c \).

Our films were prepared by molecular-beam epitaxy on 12.7 mm \( \times \) 12.7 mm \( \times \) 0.35 mm SrTiO\(_3\) substrates as detailed elsewhere \(^{[7]} \). Ce content, \( x \), is measured using inductively coupled plasma spectroscopy and is known to better than \( \pm 0.005 \). Films are highly oriented with their \( c \)-axes perpendicular to the substrate. Table I summarizes film properties. Thicknesses vary from 750 Å to 1250 Å, and one film (P6) was grown on a 250 Å buffer layer of insulating \( \text{Pr}_2\text{CuO}_4 \). \( ab \)-plane resistivities, \( \rho(T) \) (Fig. 1), are reproducible to \( \pm 15\% \). \( T_c = 23.7 \pm 0.5 \) K and transition width, \( \Delta T_c = 0.9 \pm 0.2 \) K meet or exceed values reported for nominally identical crystals \(^{[6]} \). As noted previously \(^{[7]} \), residual resistivities of PCCO films are smaller than those of PCCO crystals \(^{[8]} \). Curvature in \( \lambda(T) - \lambda(0) \) varies by more than a factor of two from crystal to crystal \(^{[6]} \). These comparisons suggest that film quality is superior to crystal quality at this time. \( \rho(T) \) in our films at 40 K is a factor of two lower than that of high-quality, comparably doped crystals of \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) (LSCO) \(^{[17]} \), the hole-doped cousin of PCCO. \( \lambda(0) = 1800 \pm 300 \) Å in our films matches the lowest value, 1930 Å, reported in LSCO near optimal doping \(^{[21]} \), and it is only slightly larger than the \( a \)-axis penetration depth, 1600 Å, in YBCO \(^{[21]} \). We therefore conclude that our films have very little disorder.

We measure \( \lambda^{-2}(T) \) with a low frequency two-coil mutual inductance technique described in detail elsewhere \(^{[22]} \). A film is centered between two small coils, and a current at about 50 kHz in one coil induces eddy currents in the film. Currents are approximately uniform through the film thickness. Data have been measured to be independent of frequency for 10 kHz \( \leq f \leq 100 \) kHz. Magnetic fields from the primary coil and the film are measured as a voltage
across the secondary coil. We have checked that the typical excitation field (100 μTesla ⊥ to film) is too small to create vortices in the film. Because the coils are much smaller than the film, the applied field is concentrated near the film’s center and demagnetizing effects at the film perimeter are irrelevant. All data presented here are in the linear response regime.

Because film thicknesses, $d$, are less than $\lambda$, it is the sheet conductivity $\sigma(\omega, T)d = \sigma_1(\omega, T)d - i\sigma_2(\omega, T)d$ that is measured, with an estimated accuracy of 5%. $\lambda^2(T)$ is obtained from $\sigma_2$: $\lambda^{-2}(T) = \mu_0\omega\sigma_2(T)$, where $\mu_0$ is the magnetic permeability of vacuum, and accuracy is limited by 5% uncertainty in $d$. The temperature dependence of $\lambda^{-2}(T)/\lambda^{-2}(0)$ is unaffected by uncertainty in $d$. The origin of 0.2% “wiggles” in $\lambda^{-2}(T)$ at low temperatures is uncertain, but we know that at least some of the effect arises from slow drift in amplifier gain.

$T_c$ and $\Delta T_c$ (Table I) are defined to be the position and full-width of the fluctuation peak in $\sigma_1(T)$ measured at 50 kHz (Fig. 2). $T_c$ from resistivity (Fig. 1) and penetration depth measurements (Figs. 2 and 3) are identical. Structure in $\sigma_1(T)$ is due to slight variation in $T_c$ through the film thickness. Our measurement technique reveals transitions in all layers of the sample, so $\Delta T_c \approx 1$ K indicates excellent film homogeneity.

Figure 3 displays $\lambda^2(T)$ in all five films (thick solid lines). The spread in $\lambda^{-2}(0)$ is larger than expected from uncertainty in $d$ [3]. Slight upward curvature in $\lambda^{-2}$ near $T_c$ is reproducible. Most importantly, the flatness of the low-temperature data is highly reproducible.

To determine the behavior of $\lambda^2(T)$ at low temperatures, we fit the first $\sim 5\%$ drop in $\lambda^{-2}(T)/\lambda^{-2}(0)$ to

$$\lambda^{-2}(T)/\lambda^{-2}(0) \sim 1 - C_\infty e^{-D/t},$$

with $t = T/T_c$. $D$ is roughly the minimum gap value, $\Delta_{min}$, normalized to $T_c$ and is fixed at 0.85 for all films to emphasize sample-to-sample reproducibility. $C_\infty$ and $\lambda^{-2}(0)$ (Table I) are adjusted for each film to yield the fits (dotted curves) in Fig. 4. The exponential curves in Fig. 4 are not best fits, but their $\chi^2$ values (Table I) lie within the estimated experimental noise of $\pm 0.2\%$ (error bars in Fig. 4). The worst fits are improved by a factor of two by adjusting $D$ by 10%. We note that a more thorough analysis finds that the peak in the density of states is near 2.2 $\mu B T_c$ in optimally doped PCCO [2].

For comparison, quadratic and cubic fits of the form $\lambda^2(T) \sim \lambda^{-2}(0) [1 - (T/T_0)^x]$ with $x = 2$ or 3, were also performed. They are shown for film P2 in Fig. 4. For all films, the best quadratic fits are visibly poorer than cubic or exponential fits. $\chi^2$ values for quadratic fits are typically twice as large as for exponential fits. We note that cubic fits possess the smallest $\chi^2$ values, leaving open the possibility that the superconducting density of states increases as energy cubed at low energies rather than being fully gapped [2]. No $d$-wave theory predicts such behavior.

Figure 3 displays $\lambda^2(T)$ from 0 K to $T_c$ along with low-$T$ exponential fits from Fig. 4 and low-$T$ quadratic and cubic fits extrapolated to higher temperatures. Exponential fits are cut off at 8 K, since the temperature dependence of the gap is neglected in the low-$T$ expression, Eq. (1). We have already demonstrated that low-$T$ quadratic fits are unacceptably poor. Even if one believes that the low-temperature data are quadratic in $T$, there is no sign of a crossover to linear behavior [4], and the extrapolated low-$T$ quadratic go to zero well above $T_c$, so the curvature is too weak to be compatible with a dirty $d$-wave interpretation [13]. We note that previous observations of quadratic behavior in $\lambda(T)$ [4] do not find a crossover from quadratic to linear, weakening their support for $d$-wave superconductivity.

In conclusion, we have presented high-precision measurements of $\lambda^{-2}(T)$ in five optimally doped PCCO films. Resistivities, $T_c$’s, transition widths, and superfluid densities are reproducible and indicative of superior film quality. In every film, $\lambda^{-2}(T)$ at low temperatures is flatter than $T^2$ and is well fitted by an exponential temperature dependence with the same minimum gap value of 0.85 $\mu B T_c$, indicating a nodeless gap in optimally doped PCCO.

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FIG. 1. $ab$-plane resistivities, $\rho(T)$, of five optimally doped PCCO films. Inset displays transition region for a typical film, P2.
FIG. 2. $\sigma_1(T)$ at 50 kHz for five optimally doped PCCO films. $T_c$ and $\Delta T_c$ are the temperature and full-width of the fluctuation peak in $\sigma_1$. For comparison, $\sigma_2(T)$ is shown for film P3.

FIG. 3. $\lambda^{-2}(T)$ (thick lines) for the ab-plane of five optimally doped PCCO films. Successive curves are offset by 10 $\mu m^{-2}$ for clarity. Thin solid (dashed) lines are extrapolations of the best cubic (quadratic) fits to low-$T$ data. Dotted curves are exponential fits to low-$T$ data, and $t \equiv T/T_c$. See Fig. 4 and text for explanation of low-$T$ fits.
FIG. 4. First ~ 5% drop in $\lambda^{-2}(T)/\lambda^{-2}(0)$ for five optimally doped PCCO films. Successive curves are offset by 0.02 for clarity. Dotted curves are fits to this data of $1 - C_\infty e^{-D/t}$, with $t \equiv T/T_c$ and $D = 0.85$. The fits lie within the experimental noise of 0.2%, represented by error bars. The thin solid (dashed) line is a cubic (quadratic) fit, described in the text. Best-fit quadratics lie outside the experimental noise and are therefore unacceptable.

TABLE I. PCCO film properties. $d$ is film thickness. $T_c$ and $\Delta T_c$ are the temperature and full-width of the fluctuation peak in $\sigma_1(T)$. Uncertainty in $d$ and $\lambda^{-2}(0)$ are estimated to be ±5%. $\rho(30 \text{ K})$ is the $ab$-plane resistivity just above $T_c$. $C_\infty$ is a fit parameter from Eq. (1). $\chi^2 \equiv (1/N) \sum [\lambda^{-2}(T) - \lambda^{-2}_f(T)]^2/\lambda^{-2}_f(T)^2$, where $N =$ number of data points, is a measure of exponential fit quality; $\chi^2$ from $T^2$ fits are in every case poorer.

| Film | $d$ [Å] | $T_c$ [K] | $\Delta T_c$ [K] | $\lambda(0)$ [Å] | $\rho(30 \text{ K})$ [$\mu\Omega$ cm] | $C_\infty$ | $\chi^2 \times 10^{-6}$ |
|------|--------|-----------|------------------|------------------|-------------------------------|--------|------------------|
| P2   | 750    | 23.2      | 0.7              | 2100             | 21                            | 0.68   | 2.56             |
| P3   | 1000   | 24.2      | 1.0              | 1800             | 19                            | 0.93   | 3.69             |
| P4   | 1250   | 23.9      | 0.7              | 1800             | 18                            | 0.78   | 2.30             |
| P5   | 750    | 23.2      | 1.1              | 2200             | 23                            | 0.79   | 4.86             |
| P6   | 750    | 23.6      | 0.8              | 1500             | 26                            | 0.69   | 3.22             |