Magnetic Penetration Depth Measurements of Pr$_{2-x}$Ce$_x$CuO$_4$–δ Films on Buffered Substrates: Evidence for a Nodeless Gap

Mun-Seog Kim, John A. Skinta, and Thomas R. Lemberger
Department of Physics, Ohio State University, Columbus, OH 43210-1106

A. Tsukada and M. Naito
NTT Basic Research Laboratories, 3-1 Morinosato Wakamiya, Atsugi-shi, Kanagawa 243, Japan

We report measurements of the inverse squared magnetic penetration depth, $\lambda^{-2}(T)$, in Pr$_{2-x}$Ce$_x$CuO$_4$–δ (0.115 ≤ $x$ ≤ 0.152) superconducting films grown on SrTiO$_3$ (001) substrates coated with a buffer layer of insulating Pr$_2$CuO$_4$. $\lambda^{-2}(0)$, $T_c$ and normal-state resistivities of these films indicate that they are clean and homogeneous. Over a wide range of Ce doping, 0.124 ≤ $x$ ≤ 0.144, $\lambda^{-2}(T)$ at low $T$ is flat: it changes by less than 0.15% over a factor of 3 change in $T$, indicating a gap in the superconducting density of states. Fits to the first 5% decrease in $\lambda^{-2}(T)$ produce values of the minimum superconducting gap in the range of 0.29 ≤ $\Delta_{min}/k_BT_c$ ≤ 1.01.

It is still a puzzle whether pairing symmetry in $n$-type cuprates is $d$ wave or not. Recently, novel concepts on pairing symmetry of $n$- and $p$-type cuprates have come forward: a possible transition in pairing symmetry[12, 13, 14] and/or a mixed symmetry order parameter[15]. Our previous work[10] in-
drive current. The absolute accuracy of $\lambda^{-2}$ is limited by $\pm 10\%$ uncertainty in $d$. The $T$ dependence of $\lambda^{-2}$ is unaffected by this uncertainty.

Except for differences in the flatness of $\lambda^{-2}$ at low $T$, which is the focal point of this paper, buffered films are very much like unbuffered films reported earlier. Fig. 1 shows in-plane resistivity, $\rho_{ab}(T)$, for buffered PCCO films. $\rho_{ab}$ in the normal state decreases smoothly and monotonically with Ce doping, $x$, even for small changes in $x$, implying that the main difference among films is Ce content. If there were random variations in degree of epitaxy, structural defects, etc., then resistivity would not be such a smooth function of $x$. These resistivities are slightly smaller than for PCCO films without buffer layers, and significantly lower than for NCCO and PCCO crystals. The inset of Fig. 1 shows that resistive transitions are reasonably sharp, and that $T_c$ is a weak function of Ce concentration, although resistivity is not. Table I summarizes properties of the films.

Fluctuations cause $\sigma_1(T)$ to peak at the superconducting transition. Hence, $\sigma_1(T)$ is a much more stringent test of film quality than resistivity. For example, if $T_c$ varies through the film thickness, resistivity reveals only the highest $T_c$. Because our probing magnetic field passes through the film, $\sigma_1(T)$ has a peak at the $T_c$ of every layer. Transitions associated with small bad spots in the film, as opposed to an entire film layer, are distinguished by their having essentially no effect on the superfluid response, $\sigma_2$. When a layer goes superconducting there is a distinct change in the slope of $\lambda^{-2}(T)$.

$\sigma_1(T)$’s of buffered PCCO films (Fig. 2) show that several of them have a double transition, reflected as shoulder ($x = 0.115, 0.124$, and 0.137) or satellite ($x = 0.144$ and 0.152) structure of peaks. We define two transition temperatures, $T_{c1}$ and $T_{c2}$, from peaks in $\sigma_1(T)$, where $T_{c1} > T_{c2}$. The resistive $T_c$ is always at the onset of the $T_{c1}$-peak. For the films most important to the conclusions of this paper, $0.124 \leq x \leq 0.144$, the width of the $T_{c1}$ peak, $\Delta T_{c1}$, is $\leq 1$ K, indicating excellent film homogeneity. The peak at $T_{c2}$ most likely involves a bad spot in the film, since there is no corresponding feature in the slope of $\lambda^{-2}(T)$, (see Fig. 3). Accordingly, the lower transition is neglected in our analysis. Films with highest and lowest Ce concentrations ($x = 0.115$ and 0.152) have broader transition widths ($\Delta T_{c2} = 2.4 \sim 3.9$ K) than other films, perhaps because $T_c$ is more sensitive to $x$.

Figure 3 shows $\lambda^{-2}(T)$ for all films. $\lambda^{-2}(0)$ vs. $x$ increases rapidly for $x \leq 0.133$, and it is constant or decreases slowly for $x > 0.133$. Values of $\lambda^{-2}(0)$ are slightly higher than for unbuffered films. The surprising upward curvature that develops in $\lambda^{-2}(T)$ near $T_c$ at high Ce concentrations was also observed in unbuffered LCCO and PCCO films.

In our previous work on unbuffered PCCO films,
films with low Ce concentrations showed quadratic ($T^2$) behavior in $\lambda^{-2}(T)$ at low $T$. Films with high Ce concentrations showed gap-like behavior:

$$\lambda^{-2}(T) \approx \lambda^{-2}(0)[1 - C_\infty \exp(-D/t)],$$

(1)

where $C_\infty$ and $D$ are adjustable parameters, and $t = T/T_c$. In the clean limit, $D$ is approximately the minimum gap on the Fermi surface, normalized to $k_BT_c$, and $C_\infty$ is roughly twice the average superconducting density of states (DOS) over energies within $k_BT$ of the gap edge. For isotropic BCS superconductors, the best-fit value of $C_\infty/2$ is about 2.2. The change in low-$T$ behavior of $\lambda^{-2}(T)$ near optimal doping suggested a transition in pairing symmetry.

We now turn to the low-$T$ behavior of $\lambda^{-2}(T)$ for buffered PCCO films, shown on a greatly expanded scale in Fig. 4. The most important thing to notice is that $\lambda^{-2}(T)$ is flat to better than 0.15% over a factor of 3 or more change in $T$. Residual variations in $\lambda^{-2}(T)$ at the 0.1% level are due, at least in part, to slow drift in the gain of the lock-in amplifiers used to measure current and voltage. These data are incompatible with simple $d$-wave models with nodes in the gap. Thus, except for the most underdoped and overdoped films ($x = 0.115$ and 0.152), $\lambda^{-2}(T)$ shows gapped behavior. Recent angle-resolved photoemission spectroscopy measurements indicate well-defined quasiparticle states on the Fermi surface where the $d_{x^2-y^2}$ node would be, so the gapped behavior that we observed could not be ascribed to a Fermi surface effect.

To estimate the gap, we fit Eq. (1) to the first ~5% drop in $\lambda^{-2}(T)$, (thin solid lines in Fig. 4). It comes as no surprise that quadratic fits over the same temperature range are unacceptable (dashed lines). For films with $x = 0.115$ and 0.152, data at $T < 0.5$ K are needed to distinguish between $T^2$ and $e^{-D/t}$. Values of the minimum gap, $\Delta_{\min} = Dk_BT_c$ and average DOS, $C_\infty/2$, extracted from the above exponential fits are presented in Table I. $D$ values are significantly lower than the BCS weak-coupling-limit value, 1.76, for $s$-wave superconductors (2.14 for $d$-wave superconductors). $D$ is largest, $D \sim 1$, for $x$ near 0.13. A similar value, $D \approx 0.85$, was found for unbuffered PCCO films with the same Ce concentration. Values of $C_\infty/2$ ($\ll 1$) are also much smaller than for weak-coupling isotropic $s$ wave. This implies existence of a peak in the DOS for a certain $E$ ($> \Delta_{\min}$), because the states should be conserved.

The next question is: where is the peak in the DOS, i.e., how big is the maximum gap, $\Delta_{\max}$, on the Fermi surface? To answer this question, we employ a model anisotropic gap function and the clean-limit result that $1 - \lambda^{-2}(T)/\lambda^{-2}(0)$ is an integral of quasiparticle DOS times the derivative of the Fermi function with respect to energy $\lambda$. Fig. 4 shows a good fit to $\lambda^{-2}(T)$ for film with $x = 0.131$ using the DOS in the inset. In this fit, the minimum gap was fixed at the value found by fitting the low-$T$ data, i.e., $\Delta_{\min}/k_BT_c = 0.99$. Then, as one can see in inset of Fig. 4, the average DOS within $\sim k_BT$ of the minimum gap edge agrees well with $C_\infty/2 = 0.5$ from Table I. For film with $x = 0.131$, $\Delta_{\max}$ is about 2.6$k_BT_c$ ($\pm 15\%$).

We emphasize that we cannot say anything about the shape of the peak in the DOS, only its location. An equally acceptable fit, with a similar peak energy, is obtained even when the sharp narrow peak in the inset of Fig. 4 is replaced by a rectangular peak.

In summary, we measured the inverse squared magnetic penetration depth, $\lambda^{-2}(T)$, of several Pr$_{2−δ}$Ce$_x$CuO$_{4−δ}$ films on buffered Pr$_2$CuO$_4$/SrTiO$_3$ substrates down to $T/T_c < 0.03$. Overall, the resistivities and penetration depths were similar to films grown directly on SrTiO$_3$. However, for PCCO films on buffered substrates, $\lambda^{-2}(T)$ at low $T$ exhibits gapped behavior over a wide range of Ce doping, including underdoping. This implies a superconducting gap without nodes on the Fermi surface. Values of the minimum superconducting gap for the films are in range of 0.3 $\leq \Delta_{\min}/k_BT_c \leq 1.0$. We cannot distinguish among models with various gap symmetries, e.g., anisotropic $s$, $s + id$, or $d + id$.

The research at OSU was supported by NSF Grant No. DMR-0203739.
Buffered Pr$_{2-x}$Ce$_x$CuO$_4$ ($x = 0.131$)

FIG. 5: $\lambda^{-2}(T)$ for Pr$_{1.869}$Ce$_{0.131}$CuO$_4-\delta$ film. Gray line shows an excellent fit obtained with density of states shown in the inset. Inset: Quasiparticle density of states in $s + id_{x^2-y^2}$ gap symmetry.

TABLE I: Properties of eight MBE-grown Pr$_{2-x}$Ce$_x$CuO$_4-\delta$ films on Pr$_2$CuO$_4$/SrTiO$_3$. $T_c$ (or $T_{c1}$) and $T_{c2}$ are locations of main and secondary peaks in $\sigma_1(T)$, respectively. $\Delta T_c$ is full width of the (main) peak in $\sigma_1(T)$. $\rho_{ab}(25\text{ K})$ is the $ab$-plane resistivity at $T = 25\text{ K}$. $\lambda^{-2}(0)$, $C_{\infty}/2$, and $D = \Delta_{\text{min}}/k_B T_c$ are fit parameters, in Eq. (1).

| $x$   | $T_c$ ($T_{c1}$) | $T_{c2}$ | $\Delta T_c$ | $\rho_{ab}(25\text{ K})$ | $\lambda^{-2}(0)$ | $C_{\infty}/2$ | $D$    |
|-------|-----------------|----------|--------------|--------------------------|------------------|-------------|--------|
|       | (K)             | (K)      | (K)          | ($\mu\Omega\text{cm}$)  | ($\mu\text{m}^{-2}$) |             |        |
| 0.115 | 13.0            | 11.8     | 3.9          | 51.0                     | 6.6              | 0.21        | 0.29   |
| 0.124 | 21.3            | 20.7     | 1.3          | 30.1                     | 19.1             | 0.28        | 0.56   |
| 0.127 | 23.1            | 23.1     | 0.5          | 18.4                     | 25.8             | 0.60        | 1.01   |
| 0.132 | 23.5            | 23.6     | 0.5          | 18.3                     | 27.9             | 0.50        | 0.99   |
| 0.133 | 23.3            | 23.3     | 0.5          | 15.3                     | 27.9             | 0.50        | 0.99   |
| 0.137 | 23.2            | 22.9     | 0.7          | 10.8                     | 41.2             | 0.38        | 0.83   |
| 0.144 | 21.2            | 20.2     | 0.9          | 9.5                      | 38.6             | 0.30        | 0.72   |
| 0.152 | 19.8            | 16.6     | 2.4          | 7.7                      | 35.1             | 0.17        | 0.37   |

[1] C. C. Tsuei and J. R. Kirtley, Phys. Rev. Lett. 85, 182 (2000).
[2] N. P. Armitage et al., Phys. Rev. Lett. 86, 1126 (2001).
[3] J. D. Kokales et al., Phys. Rev. Lett. 85, 3696 (2000).
[4] R. Prozorov, R. W. Giannetta, P. Fournier, and R. L. Greene, Phys. Rev. Lett. 85, 3700 (2000).
[5] L. Alff et al., Phys. Rev. B 58, 11197 (1998).
[6] S. Kashiwaya et al., Phys. Rev. B 57, 8680 (1998).
[7] C.-T. Chen et al., Phys. Rev. Lett. 88, 227002 (2002).
[8] L. Alff et al., Phys. Rev. Lett. 83, 2644 (1999).
[9] J. A. Skinta, T. R. Lemberger, T. Greibe, and M. Naito, Phys. Rev. Lett. 88, 207003 (2002).
[10] J. A. Skinta et al., Phys. Rev. Lett. 88, 207005 (2002).
[11] A. Biswas et al., Phys. Rev. Lett. 88, 207004 (2002).
[12] A. Kohen and G. Deutscher, cond-mat/0207382 (2002).
[13] K. A. Müller, Phil. Mag. Lett. 82, 279 (2002).
[14] D. Daghero, R. S. Gonnelli, G. A. Ummarino, and V. A. Stepanov, cond-mat/0207411 (2002).
[15] M. Naito, H. Sato, and H. Yamamoto, Physica C 293, 36 (1997).
[16] S. J. Turneaure, E. R. Ulm, and T. R. Lemberger, J. Appl. Phys. 79, 4221 (1996).
[17] S. J. Turneaure, A. A. Pesetek, and T. R. Lemberger, J. Appl. Phys. 83, 4334 (1998).
[18] J. D. Kokales et al., Physica C 341-348, 1655 (2001).
[19] M. Tinkham, Introduction to Superconductivity, 2nd ed. (McGraw-Hill, New York, 1996).
[20] J. A. Skinta et al., cond-mat/0301174 (2003).