Magnetic penetration depth in superconducting $La_{2-x}Sr_xCuO_4$ films

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We have measured the magnetic penetration depth $\lambda(T)$ in a series of superconducting $La_{2-x}Sr_xCuO_4$ films with Sr concentrations from $x=0.135$ to 0.175. $\lambda^{-2}(T)-\lambda^{-2}(0)$ is quadratic in T for $0.4<T<10$ K, which puts an upper limit of about 4 K on a possible isotropic gap. The larger magnitude of $\lambda(0)$, reduced T_C, and higher resistivity of films relative to bulk samples, plus the T^2 behavior of $\lambda^{-2}(T)-\lambda^{-2}(0)$, lead naturally to the conclusion that superconductivity in $La_{2-x}Sr_xCuO_4$ is d wave. Near T_C the real part of the conductivity and $\lambda(T)$ are analyzed for critical behavior. The data indicate that the critical region is no wider than 1 K. [S0163-1829(98)02646-0]

I. INTRODUCTION

A fundamental question in the high-T_C cuprate superconductors is the symmetry of the order parameter, which may vary among the cuprates despite the commonality of quasi-two-dimensional copper-oxide planes. There is a strong consensus that $YBa_2Cu_3O_{7-\delta}$ (Refs. 1–3) (YBCO) and $Bi_2Sr_2CaCu_2O_8$ (Refs. 4 and 5) (BSCCO) have d-wave order parameters, but there is evidence that $Nd_{2-x}Ce_xCuO_4$ (NCCO) is an s-wave superconductor.[6–10] The question remains whether $La_{2-x}Sr_xCuO_4$ manifests d- or s-wave symmetry. $La_{2-x}Sr_xCuO_4$'s crystal structure and T_C are similar to those of NCCO. However, $La_{2-x}Sr_xCuO_4$ is hole doped like YBCO and BSCCO while NCCO is electron doped. Previous studies of Raman spectra,[11] infrared reflectance,[12] neutron scattering, and heat capacity[13] of $La_{2-x}Sr_xCuO_4$ have found results that are inconsistent with a simple isotropic s-wave gap. The present paper examines the question through analysis of the magnetic penetration depth, $\lambda(T)$. Penetration depth studies have proven to be critically important in understanding the order-parameter symmetry in other compounds, because unlike other experimental quantities, there is no normal-state background associated with $\lambda(T)$.

The magnetic penetration depth reflects the symmetry of the order parameter primarily through its dependence on T at $T/T_C \ll 1$. The distinction between d-wave and s-wave symmetries is well defined and experimentally accessible, particularly when there is some disorder in the sample. For an anisotropic superconductor with a nonzero minimum energy gap Δ_{min} in its excitation spectrum, $\lambda^{-2}(T)$ is clearly exponential, $\lambda^{-2}(T)-\lambda^{-2}(0)\sim e^{-\Delta_{min}/kT}$, for kT less than about $0.1\Delta_{min}$. For an isotropic BCS superconductor with a square-root singularity in its density of states at $E=\Delta$, exponential behavior persists up to about 0.3Δ. In this case the exponential behavior is not affected by nonmagnetic disorder or by small levels of magnetic disorder, although the magnitude of Δ generally is. In fact, if the s-wave gap is anisotropic, then disorder tends to make it isotropic, thereby increasing Δ_{min} and making the exponential behavior in $\lambda^{-2}(T)$ more prominent. Small levels of magnetic disorder smear out any singularity in the density of states and reduce Δ.

For a d-wave order parameter, $\lambda^{-2}(T)$ is linear in T, $\lambda^{-2}(T)-\lambda^{-2}(0)\propto T$, for very clean samples, and it is quadratic, $\lambda^{-2}(T)-\lambda^{-2}(0)\propto T^2$, for disordered samples.[14] Only a very few pure YBCO (Ref. 3) and BSCCO (Refs. 5 and 15) single crystals exhibit a linear low-T penetration depth, presumably due to the sensitivity of the density of states to small amounts of disorder. Most crystals show quadratic behavior, even nominally excellent crystals with very sharp transitions and low resistivities. None show gaplike behavior. By contrast, in three NCCO films λ is reported to vary exponentially, $\lambda^{-2}(T)-\lambda^{-2}(0)\sim e^{-\Delta/kT}$.[8–10]

Since the present paper deals with $La_{2-x}Sr_xCuO_4$ films, without the benefit of crystals for comparison, it is germane to note how YBCO films compare with YBCO crystals. Some films made by codeposition of Y, BaF_2, and Cu with a high-temperature postanneal in flowing oxygen show linear behavior of λ with T from 30 K down to at least 2 K, with values of the slope and magnitude close to those that are found in crystals.[16,17] This indicates that it is possible to make films of a quality similar to that observed in crystals, despite the grain boundaries, strain due to lattice mismatch with the substrate, and disorder incorporated during deposition. In many films made by codeposition, and essentially in all films made by sputtering and by pulsed laser deposition, λ varies as T^2 below approximately 30 K, presumably resulting from the small amount of disorder incorporated during deposition.[18] There is one report of gaplike behavior in films.[19] YBCO films doped with 3–6% Ni and Zn are strongly disordered, and have greatly reduced superfluid densities, quadratic T dependencies, and reduced transition temperatures, as expected for a d-wave order parameter although T_C is not reduced as much as predicted from d-wave theory.[20,21]

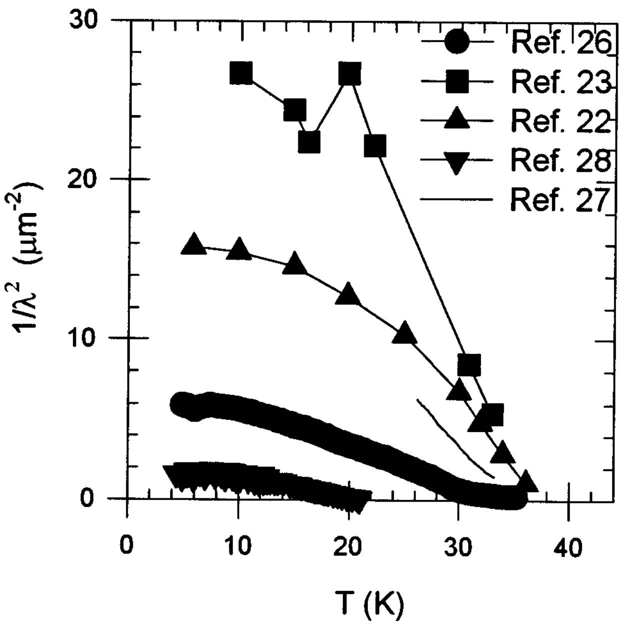

FIG. 1. $1/\lambda^2$ vs T for $La_{0.85}Sr_{0.15}CuO_4$ taken from the literature. Data are from Shibauchi *et al.* (Ref. 26), Kossler *et al.* (Ref. 23), Aeppli *et al.* (Ref. 22), Jaccard *et al.* (Ref. 28), and Li *et al.* (Ref. 27).

There are several published magnetic penetration depth studies of $La_{2-x}Sr_xCuO_4$. Results of optimally doped material ($x=0.15$) from μsr,[22–25] microwave surface impedance,[26] magnetization measurements,[27] and two-coil mutual inductance measurements[28] are shown in Fig. 1. The best low-T data are the μsr data on bulk material by Aeppli, Cava, and Ansaldo,[22] which are smooth and monotonic at low T. They show that $\lambda(T)$ is not linear over the range for which YBCO crystals are linear, namely, below $T_C/3$. However, with only three data points at low T it is not possible to differentiate between quadratic and exponential dependence. The data presented in the present paper extend to 0.4 K and have a noise level of 10 Å in $\lambda(T)$, which allows us to distinguish among exponential, quadratic, or linear T dependencies at low T. Only after years of painstaking efforts towards maximizing sample quality was it possible to observe a linear T dependence in $\lambda(T)$ for YBCO. Unless further efforts make this possible in $La_{2-x}Sr_xCuO_4$, the debate between *d*-wave and *s*-wave symmetry in $La_{2-x}Sr_xCuO_4$ is likely to be decided by distinguishing between quadratic and exponential behavior in λ.

The superconducting properties of $La_{2-x}Sr_xCuO_4$ films are different from those of bulk samples. A summary of reported values of $\lambda(0)$ and T_C for the optimally doped compound, $La_{1.85}Sr_{0.15}CuO_4$, is shown in Table I. Optimally-doped sintered pellets and crystals have values of T_C near 37 K and $\lambda(0)\approx 2500$ Å. For our films T_C ranges from 23 to 30 K and $\lambda(0)$ from 5000 to 8000 Å. Locquet and co-workers[28] reported films with $T_C=20.9$ K and $\lambda(0)\sim 7000$ Å. The differences between films and bulk materials are attributed to disorder, strain, and oxygen vacancies. Since disorder is expected to enhance gaplike behavior in anisotropic *s*-wave superconductors, disorder makes it easier to distinguish *d*-wave from *s*-wave behavior.

II. EXPERIMENT

The experiment involves measurement of the complex mutual inductance of two counterwound coils that are coaxial, with the film centered between them.[29,30] The coils are pressed against the film so that the space between the coils is minimized and the signal maximized. The in-phase and out-of-phase components of the mutual inductance are inverted to obtain the complex conductivity, $\sigma=\sigma_1-i\sigma_2$. There are no adjustable parameters. The inversion calculation includes the variation of the fields and currents through the thickness of the film.[31,32] As explained in Ref. 30, the inversion procedure involves subtracting a calibration constant equal to the mutual inductance when the film is replaced by a thick superconducting Pb foil with the same shape as the film. For small films the uncertainty in the calibration constant becomes significant because of the uncertainty in locating the film and the foil in precisely the same spot. It is important to emphasize that this uncertainty does not affect our result that $\lambda^{-2}(T)-\lambda^{-2}(0)$ is quadratic in T at low temperatures. Figure 2 illustrates the full range of variation in the experimental $\lambda(T)$ from the uncertainty in the calibration constant for a typical film.

TABLE I. Summary of previous measurements of λ in optimally doped $La_{2-x}Sr_xCuO_4$.

Ref.	Experiment	Sample	T_C ($x=0.15$) (K)	$\lambda(0)$ (Å)
Uemura *et al.* (Ref. 24)	μsr	sintered pellets	37 30	2040 2700
Kossler *et al.* (Ref. 23)	μsr	sintered pellets	37	2000
Aeppli *et al.* (Ref. 22)	μsr	sintered pellets	37	2500
Frank *et al.* (Ref. 25)	μsr	sintered pellets	38	2200
Locquet *et al.* (Ref. 28)	two-coil method	films	22	7000
Shibauchi *et al.* (Ref. 26)	microwave surface impedance	single crystal	34.9	3000

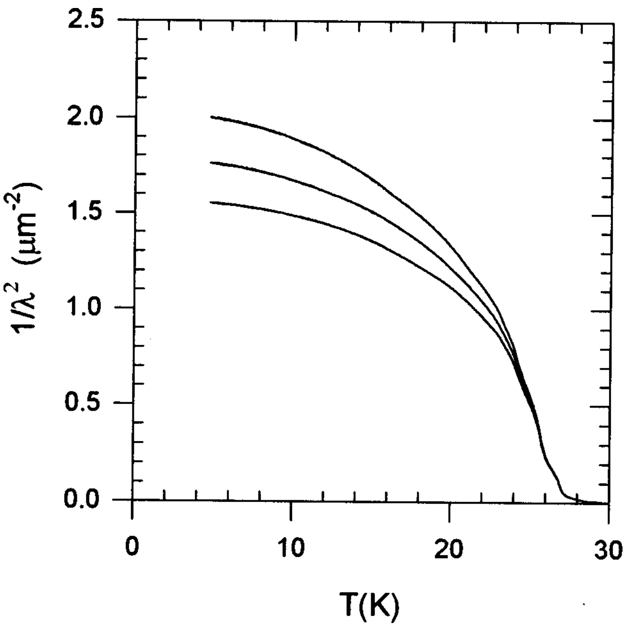

FIG. 2. $1/\lambda^2$ vs T for $La_{1.85}Sr_{0.15}CuO_4$ film $B3$ generated from one measurement but with subtraction of different calibration constants, all lying within the experimental error bars. $\lambda(4\ K)=7500\pm500$ Å.

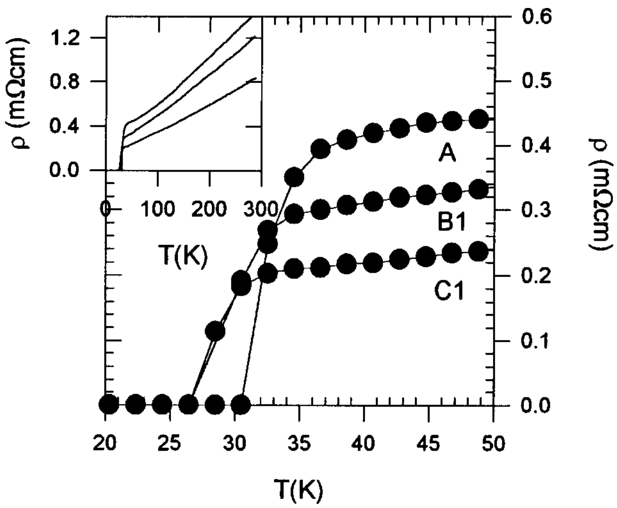

FIG. 3. Resistivity vs T for $B1$ ($x=0.15$), A ($x=0.135$), and $C1$ ($x=0.175$).

III. SAMPLE PREPARATION

Thin films of $La_{2-x}Sr_xCuO_4$ were made by pulsed laser deposition. Details have been published elsewhere.[33,34] Four films were about 1000 Å thick. In an attempt to reduce the effects of strain, five films were grown with thicknesses from 2000 to 5000 Å. In all of the films λ is greater than the film thickness, so that the measurement probes through the entire film. All of the films were deposited on either 5- or 7-mm-square $LaSrAl_2O_4$ substrate, which have an excellent lattice match to the films. Three of the films, A, $B1$, and $C1$, were postannealed at 400 °C in 70 bars of O_2 to increase their oxygen concentration.[34] This procedure increased T_C by a few degrees.

After the mutual inductance measurements the films were patterned into 4 mm by 2-mm strips for resistivity measurements. The resistivities of the postannealed films are shown in Fig. 3. Their resistivities at 300 K are about four times as large as for bulk $La_{2-x}Sr_xCuO_4$ as measured by Takagi *et al.*, who report $\rho(300\ K)=0.24$ mΩ cm for $x=0.15$.[35] The residual resistivity also indicates higher disorder in the films than in crystals. X-ray diffraction of similar films shows excellent c-axis alignment with the substrate and an absence of foreign phases.

IV. PENETRATION DEPTH RESULTS

We have measured the penetration depth in six optimally doped films with nominal Sr concentrations of $x=0.15$, and values of T_C between 20 and 30 K, one underdoped film with $x=0.135$ and $T_C=31$ K, and two overdoped films with $x=0.175$ and $T_C\approx27$ K (Table II). Figure 4 shows the temperature dependence of $1/\lambda^2$, which is proportional to the superfluid density. Figure 5 shows the same data, scaled so as to overlap in the low-T region. The two thickest films ($B2$ and $C2$) have the sharpest transitions. The peak in σ_1 at T_C for each of these films shows a full width at half-maximum of about 0.8 K. $1/\lambda^2$ near T_C for these two specimens is shown in Fig. 6.

Although the postannealed optimally doped specimen (B1) has the highest superfluid density, all three postannealed specimens (A, $B1$, and $C1$) have broad transitions, with C1 showing distinct multiple transitions. These features, together with the fact that T_C was only slightly increased by the annealing procedure, indicate that the oxygenation was incomplete, so that different layers have different values of T_C.

The four remaining films are optimally doped and thinner. LSCO films of that thickness do not exhibit any superconductivity when deposited on $SrTiO_3$, and only the use of a substrate whose lattice parameter is better matched to the film allows us to study them at all.[33] However, the lattice match is still not perfect, and some effect of the residual strain may be expected. The fact that these four films do not follow a singe pattern illustrates the limited reproducibility. The energy of the laser pulses is not quite constant. Small changes in the position of the substrate with respect to the plume of material produced by the laser pulse can cause substantial changes. The present results indicate that it is likely to be possible to improve film quality by further reduction of lattice mismatch and by careful selection of speci-

TABLE II. Summary of samples reported in this paper. Sr concentrations are nominal.

Film name	"x" in $La_{2-x}Sr_x$	T_C (K)	λ (T=4 K) (Å)	Thickness (Å)
A	0.135	31	5900±300	3600
$B1$	0.15	30.5	4500±300	2700
$B2$	0.15	27.5	6000±2000	5000
$B3$	0.15	25–30	6600±500	1200
$B4$	0.15	27	7400±500	1120
$B5$	0.15	23	7600±500	1200
$B6$	0.15	25	8400±500	900
$C1$	0.175	20–27	6000±300	3400
$C2$	0.175	27.5	7000±2000	5000

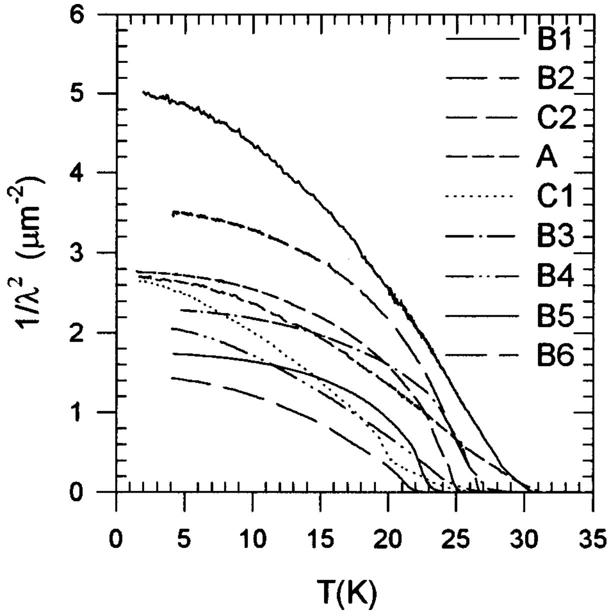

FIG. 4. $1/\lambda^2(T)$ vs T for all nine films in this study. The labels for the curves are arranged in order of decreasing superfluid density at 1 K.

mens. The films in this study were entirely unselected. Further improvements in T_C are also possible, as illustrated recently by Sato *et al.*[36]

In spite of the variations in film thickness, T_C, sharpness of the transition, and superfluid density, the temperature dependence of $1/\lambda^2$ is the same for all films at low T as shown in Fig. 5. This is our central result, namely that the temperature dependence of λ^{-2} is the same for all films at low T, and is quadratic. Figure 7 shows quadratic fits to the data for films A, $B1$, and $C1$. We show these three films since they have the largest superfluid density for their respective Sr concentrations, and their sample dimensions provided the most accuracy in measuring their superfluid density. More recently we have been able to extend the measurement to 0.4 K and data on film $B6$ are shown in Fig. 8 for this extended temperature range. The quadratic fits to A, $B1$, and $C1$ are

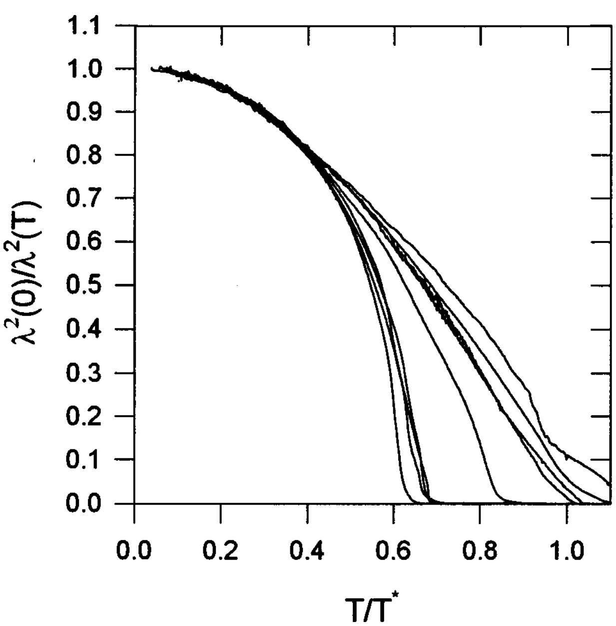

FIG. 5. $1/\lambda^2(T)$ vs T, normalized so that the data agree at low T. The upper data sets are for films A, $B1$, $B4$, $B6$, and $C1$. The lower data sets are for films $B2$, $B3$, $B5$, and $C2$. The data clearly show two functional forms.

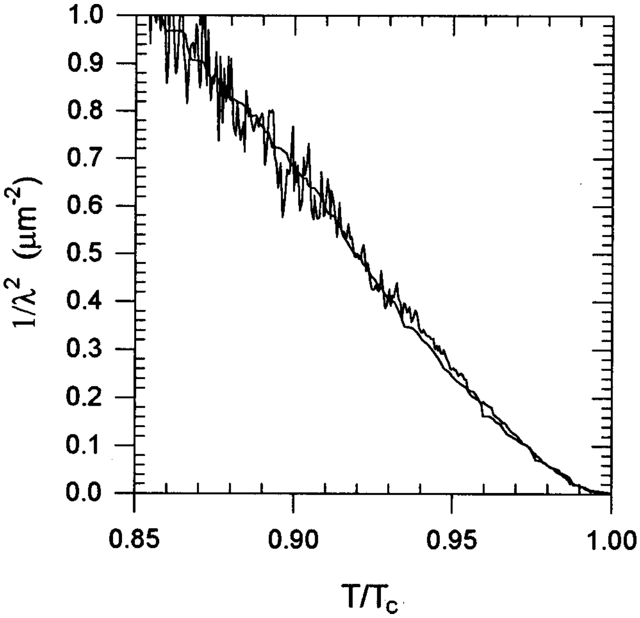

FIG. 6. $1/\lambda^2$ vs T for films $B2$ and $C2$ near $T_C = 27.5$ K.

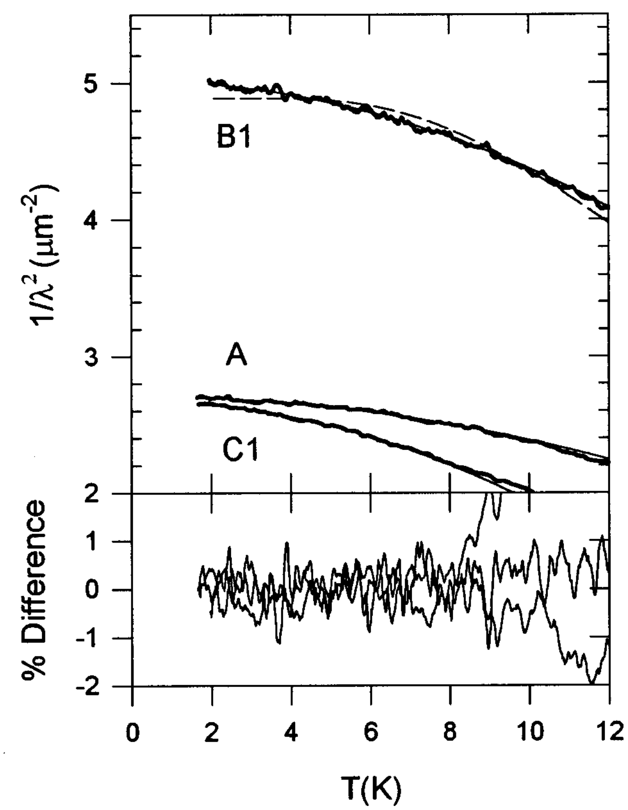

FIG. 7. Quadratic fits to $\lambda^{-2}(T)$ below 10 K. Data are shown for films $B1$ ($x = 0.15$), A ($x = 0.135$), and $C1$ ($x = 0.175$). The dashed line is a fit to $B1$ of a BCS temperature dependence with $2\Delta/kT_C = 2.6$.

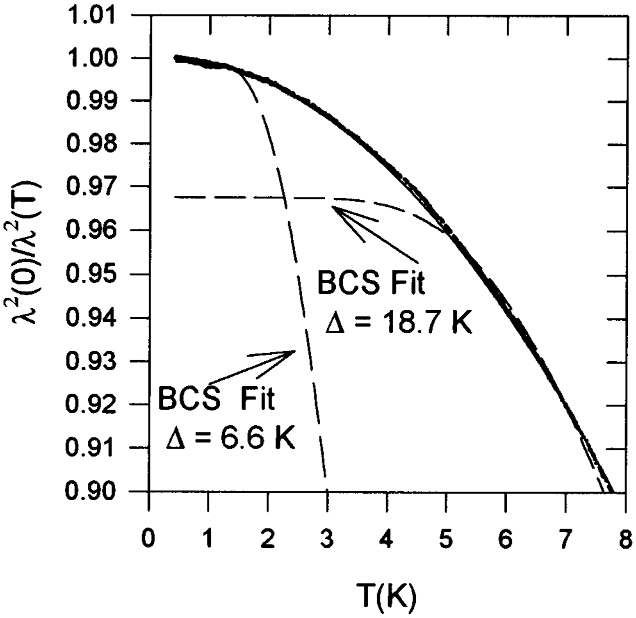

FIG. 8. Quadratic fit for $\lambda^2(0)/\lambda^2(T)$ for film $B6$. Note the expanded vertical scale. The BCS best fit to the data between 4 and 8 K is shown with a gap of 18 K, as well as a BCS best fit to the data from 0.4 to 2 K with a gap of 6 K.

within $\pm 1\%$ of the data. A fit to the standard BCS temperature dependence for λ^{-2} is also shown for film B1 in Fig. 7 and the error for this fit is greater than 2%. The value for $2\Delta/kT_C$ from the best fit is 2.6, which is less than the BCS weak-coupling value of 3.5 and less than the value of 4.1 found in NCCO.[7] With the lower temperature data of $B6$ it is clear from Fig. 8 that a gap of this magnitude will not fit the lowest-temperature data. In fact, the only range over which a BCS temperature dependence might be argued to fit is from 0.4 to 2 K. This fit is shown for film B6 and gives $2\Delta/kT_C = 0.9$. A quadratic temperature dependence fits better than an exponential down to the lowest temperature measured. Given that the resistivity indicates a substantial level of disorder in these films, these data strongly support the conclusion that $La_{2-x}Sr_xCuO_4$ is a d-wave superconductor.

The magnitude of $\lambda(0)$ in these films is much larger than the value, 2500 ± 400 Å, which has been reported for crystals and sintered pellets. The most convenient quantity for comparison with d and s-wave models is the superfluid density, $n_s(0)\propto\lambda^{-2}(0)$. Among our films, $n_s(0)$ is smaller than in bulk materials by factors of 4–10. In a d-wave interpretation this is a natural consequence of disorder competing with an anisotropic gap that cannot be made isotropic by disorder. However, the theory also predicts a decrease in T_C by the same factor as the decrease in $n_s(0)$ which is not observed.[20] In YBCO samples disordered by chemical substitution of the Cu ions a similar behavior of $\lambda(0)$ is observed.

Another possible explanation for the increase in λ is that the measured inductance is dominated by grain boundaries. If a grain boundary is modeled as a resistively shunted Josephson junction then the effective λ would be given by $\lambda_{\rm eff}^2=\lambda^2+\lambda_J^2$ where $\lambda_J^2=\hbar d/2eI_c\mu_0$.[37] The smaller the critical current, I_c, of the Josephson junctions the larger their contribution to λ. It is not possible to separate the contribution from the grains and the grain boundaries out of hand. We have neglected grain boundary Josephson junctions in our analysis.

V. CRITICAL REGION

A key issue for high-T_C cuprates is the temperature range near T_C where critical fluctuations dominate. In two pure YBCO crystals reported in the literature,[38] $\lambda^{-2}(T)$ shows $3d$-XYcritical behavior, $\lambda^{-2}(T)\propto(1-T/T_C)^{2/3}$, over a range of about 5 K below T_C. This is a very wide critical region. On the other hand, YBCO films consistently show mean-field behavior to within about 0.5 K of T_C.[39,40] While the details of the critical behavior are by no means clear, it is generally understood that the energy that controls the size of fluctuations is $\phi_0^2/8\pi\mu_0(\lambda^2/\ell)$, where ℓ is the coherence length in three dimensions (3D) and a sheet thickness in 2D. Thus, the large penetration depths in our $La_{2-x}Sr_xCuO_4$ films relative to $\lambda(0)\approx 1500$ Å in YBCO, are expected to result in a critical region that is at least as wide as in YBCO. In the two films with the sharpest transitions, it is clear that this does not occur. In fact, $1/\lambda^2$ curves slightly upward, rather than sharply downward, as T approaches T_C (Fig. 6). We conclude that the critical region is less than 1 K wide. We note that Locquet and co-workers[28] conclude that the critical region is about 2 K wide in their $La_{2-x}Sr_xCuO_4$ films.

VI. CONCLUSION

The characteristic features of our data on $\lambda(T)$ in $La_{2-x}Sr_xCuO_4$ films are explained in terms of a d-wave superconducting order parameter modified by the effects of the disorder generated during film fabrication. These features include the increased values of $\lambda(0)$, increased resistivities, and the suppressed values of T_C of films relative to bulk, and the quadratic T dependence of $\lambda^{-2}(T)-\lambda^{-2}(0)$ at low T. These features are shared with YBCO films deliberately disordered by chemical substitutions. Given the similarities in the structure and holelike normal-state electronic behavior of the CuO planes in YBCO and $La_{2-x}Sr_xCuO_4$, and the similarities in the behavior of $\lambda(T)$ in doped YBCO and in $La_{2-x}Sr_xCuO_4$ films, the natural conclusion is that they share the same order parameter symmetry.

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