

Absence of three-dimensional XY behavior in the magnetic penetration depth of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ films

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We have studied the transition region of numerous superconducting $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ films through measurement of the magnetic penetration depth $\lambda(T)$. Contrary to published results on $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ crystals, $\lambda(T)$ does not display three-dimensional XY behavior to within 0.5 K of T_C . Measurements are performed in negligible external magnetic fields on films made by sputtering, coevaporation, and pulsed-laser deposition. For the samples with the narrowest transitions, as determined from the width of the peak in the real conductivity $\sigma_1(T)$, the superfluid density is mean-field-like to within 0.2 K of T_C . [S0163-1829(99)07109-X]

INTRODUCTION

A thorough understanding of the role of thermal fluctuations is central to advancing a workable model of high-temperature superconductivity. Thermal fluctuations of the order parameter are pervasive, influencing essentially all measurable parameters, including the superfluid density, specific heat, complex conductivity, and magnetization. Near the superconducting-to-normal transition where the order parameter becomes vanishingly small, fluctuations are of course expected to play a prominent role. In addition, it has been proposed that thermal fluctuations play a substantial role at temperatures well away from the transition, causing significant deviations from mean-field (MF) calculations.¹⁻³ For example, it is predicted that thermal phase fluctuations will impress a linear-in- T dependence upon the superfluid density $n_S \sim \lambda^{-2}(T)$ at low temperatures. Such a contribution could mask a crossover to BCS-like behavior induced by chemical or radiation damage.

The temperature range near the transition over which fluctuations dominate behavior, known as the critical region, serves as a measure of the strength of fluctuations. The significance of low-temperature fluctuations should be in proportion to the measured strength of fluctuations near the transition. The large penetration depths λ and short coherence lengths ξ of high-temperature superconductors suggest a significant enhancement of fluctuations relative to low-temperature superconductors. Previous zero-field measurements of $\lambda(T)$ for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) single crystals indicate a critical region as wide as 5 K,^{4,5} an unexpectedly large range, suggesting remarkably strong fluctuations. In contrast to these results, measurements of $\lambda(T)$ for YBCO thin films indicate a critical region less than 1 K wide.^{6,7} Microwave measurements of the surface resistance of a crystal above the transition indicate a critical region less than 0.6-K wide.⁵ Recent measurements of $\lambda(T)$ of specially prepared, exceptionally clean YBCO single crystals show results both consistent with⁷ and inconsistent with⁸ a large critical region. The present paper is a full report of measurements on numerous YBCO films made by different techniques, having slightly different transition temperatures and zero-temperature penetration depths. Data on all films point

to the same conclusion, namely that the critical region in zero field, as determined by the behavior of $\lambda(T)$ near the transition, is less than 0.5 K, and perhaps less than 0.2 K wide.

To begin, the significance of thermal fluctuations near the transition can be estimated using the Ginzburg criterion.⁹ The Ginzburg criterion is based on a comparison of fluctuation energies to the thermal energy kT . For zero field, the predicted width of the critical region is given as

$$\Delta T \leq \frac{2\pi^2 \mu_0^2 (kT_{C0})^2}{\Phi_0^4} \left(\frac{\lambda(0)^2}{\xi(0)} \right)^2 T_{C0}, \quad (1)$$

where Φ_0 is the superconducting flux quantum, $\xi(0)$ is the Ginzburg-Landau coherence length at zero temperature, and T_{C0} is the MF transition temperature. The strong dependence on the zero-temperature penetration depth $\lambda(0)$ implies a dramatic increase in fluctuation effects as $\lambda(0)$ is increased by chemical doping or radiation damage, for example. Assuming optimistic values of $\lambda(0) = 2000 \text{ \AA}$ and $\xi = 10 \text{ \AA}$ for pure YBCO, the Ginzburg criterion predicts a critical region width $\Delta T \sim 0.4 \text{ K}$. For more conventional values of $\lambda(0) = 1400 \text{ \AA}$ and $\xi = 14 \text{ \AA}$, $\Delta T \sim 0.05 \text{ K}$. Since the Ginzburg criterion is only a rough guide, quantitatively, these low estimations do not preclude a 5 K wide critical region; however, such a broad critical region would be surprising. Measurements of fluctuation effects in conventional superconductors are experimentally inaccessible and preclude further phenomenological refinements of the Ginzburg criterion.

To further an appreciation of the ubiquitous nature of fluctuation effects, it is useful to examine their influence on the inductance of a single Josephson junction (JJ). It will also be seen that the results lead to useful parallels when considering a continuous superconducting film. Of course, there is no phase transition in a single JJ, but there is a crossover in the impedance of the device from inductive to resistive as temperature increases, just as there is in the sheet impedance of a superconducting film. The inverse of the Josephson inductance L_J is analogous to the superfluid density n_S in a continuous film. The sheet inductance of an $N \times N$ array of identical junctions is the inductance of a single junction, and the sheet inductance of a continuous thin film is $\mu_0 \lambda^2 / d \sim 1/n_S$, where d is the film thickness.

At first neglecting thermal fluctuations, when a JJ with critical current $I_C(T)$ is biased externally with a dc current I_{dc} , its effective inductance is

$$L_{J0}(T, I_{dc}) = \frac{\hbar}{2eI_C} \left[1 - \frac{I_{DC}^2}{I_C^2} \right]^{-1/2}, \quad (2)$$

where the subscript 0 indicates MF behavior. At finite temperatures, with no external bias, thermal noise currents I_S from the shunting resistance effectively bias the junction. We can estimate this mean-square ‘‘thermal bias’’ current by appealing to the equipartition theorem. That is,

$$\frac{1}{2} L_{J0} \langle I_S^2 \rangle = \frac{1}{2} kT, \quad (3)$$

or,

$$\frac{\langle I_S^2 \rangle}{I_C^2} \approx \frac{kT}{(\hbar I_C / 2e)}. \quad (4)$$

The right-hand side of the equation is recognized as the ratio of thermal energy to the Josephson coupling energy. For the sake of simplicity, we have neglected any corrections that would result when the RMS supercurrent through the junction approaches the critical current. Note that the effect of fluctuations increases as I_C decreases, in other words, as the mean-field inductance L_{J0} increases. This result agrees qualitatively with the Ginzburg criterion, i.e., ΔT increases as $\lambda^2(0)$ increases. Approximating the temperature dependence of L_J^{-1} by replacing I_{dc}^2/I_C^2 in Eq. (2) with the expression from Eq. (4),

$$\begin{aligned} L_J^{-1}(T, I_{DC}=0) &\approx L_{J0}^{-1}(T, \langle I_S^2 \rangle) \\ &\approx L_{J0}^{-1}(T, 0) \left[1 - \frac{kT}{(\hbar I_C / 2e)} \right]^{1/2}. \end{aligned} \quad (5)$$

Equation (5) demonstrates how a reduction of the Josephson inductance, or superfluid density for a continuous film, due to thermal phase fluctuations follows from a classical treatment.

Although the details are different in continuous systems, that is, there is a phase transition instead of a crossover, the same qualitative physics comes into play. In a continuous system, two other length scales become important, namely ξ , and because of its influence on λ and ξ , the electron mean free path l . Following analogous arguments for two-dimensional (2D) superconductors, with a 2D penetration depth $\lambda_{\perp} = \lambda^2/d$, the effect of thermal phase, or supercurrent, fluctuations leads to a suppression in superfluid density:

$$n_S(T) \approx n_{S0}(T) \left[1 - \frac{a(T)kT}{E_0} \right], \quad (6)$$

where the characteristic superconducting energy is

$$E_0 = \frac{\Phi_0^2}{4\pi^2 \mu_0 \lambda_{\perp}}. \quad (7)$$

The function $a(T)$ is approximately unity at all temperatures in the dirty limit $l \ll \xi$. In the clean limit $a(T)$ decreases from

unity near T_C to a small value at low T . Of course, a true 2D system will exhibit a Kosterlitz-Thouless-Berezinskii (KTB) transition with a discontinuous drop in the superfluid density;¹⁰ still, Eq. (6) captures the qualitative effects of thermal fluctuations below the KTB transition.

Moving from physical estimates to the lab, it is found that in magnetic fields of 1–5 T a rather broad critical region is clearly evident and is well described by the 3D XY model. Studies of critical dynamics in applied fields include measurements of heat capacity,^{11,12} magnetization,¹³ and I - V characteristics.^{14–16} All of these properties are found to be well described by scaling functions consistent with a 3D XY critical region several degrees wide. There is, however, no clear sense of the manner in which the width of the critical region varies as a function of field magnitude. For fields below 1 T, scaling arguments work less well, suggesting that the critical region may be much smaller. Above 5 T multicritical scaling becomes important as 3D XY behavior merges with the glassy vortex dynamics and lowest Landau-level behavior, which dominate at high fields.¹⁷ Though there are several published zero-field results^{18–20} there is no well-established consensus regarding the nature of the zero-field critical region.

The penetration depth in zero applied field serves as a powerful tool in determining the presence or absence of critical behavior for several reasons. Most importantly, because it is related directly to the superconducting order parameter, or superfluid density, it has no normal state analog, and therefore no background corrections are necessary as is the case with the real part of the conductivity and the specific heat. Also, the penetration depth can be measured with sufficient precision to permit an accurate determination of the exponent, which describes its behavior near T_C . One must be able to distinguish 3D XY behavior, where $\lambda^{-2}(T) \sim (T_C - T)^{2/3}$ from mean-field behavior, with $\lambda^{-2}(T) \sim (T_C - T)$.⁹ Finally, $\lambda(T)$ can be determined with sufficient accuracy that one can look for a correlation of fluctuation effects with the magnitude of $\lambda(0)$.

EXPERIMENT

We use a two-coil mutual inductance technique at 50 kHz to determine the complex conductivity $\sigma_1(T) + i\sigma_2(T)$ of the film from which we extract the ab -plane penetration depth. The coils are 2 mm diameter quadrupoles located on opposite sides of the film. The lateral dimensions of the films are much larger than 2 mm, typically being 1 cm square or 15 mm diameter, so the magnetic field produced by the current in the primary coil is nonuniform in both magnitude and direction over the film. The complex conductivity is extracted from the measured mutual inductance with no adjustable parameters.²¹ The mathematical calculations necessary to perform the extraction are described elsewhere.^{22–24} We perform a normalization to remove the small temperature dependence of the probe itself. We associate the imaginary conductivity σ_2 with the superfluid density in defining the penetration depth from $\lambda^{-2}(T) = \mu_0 \omega \sigma_2(T)$. This is clearly an appropriate definition when $\sigma_1 \ll \sigma_2$, and this is the case except for temperatures very close to T_C . We do not attempt to draw conclusions from data in the 0.2–0.5 K wide temperature range near T_C where $\sigma_1 \gtrsim \sigma_2$.

In the present paper, to check that the ambient field of about 0.3 G was negligible, for some samples we took data in the ambient magnetic field and with the ambient field nulled by use of an external coil. The two sets of data were essentially identical below T_C .

Great care was taken to ensure that the magnetic field from the primary coil was weak enough that any vortices it might have produced in the sample films did not affect the data. That is, we checked that data were measured in the linear response regime by measuring each sample at drive currents spanning several decades. For comparison, film behavior in the nonlinear response regime is presented (Fig. 4) and discussed below. For typical linear-response data runs, the field produced by the primary coil had a component perpendicular to the film which took its largest value, about 1 mG, at the center of the film. For the temperatures of interest here, the films were superconducting and diamagnetic, resulting in a maximum total perpendicular field a factor of 10 smaller. If H_{C1} can be approximated with the usual Ginzburg-Landau expression, $\Phi_0 \ln(\kappa)/4\pi\lambda^2$, where $1/\lambda^2$ is measured and $\ln(\kappa) \approx 5$, then H_{C1} decreases at roughly 1 G/K as T approaches T_C . Therefore, the experimental field of 1 mG should have exceeded H_{C1} only when T was within 0.1 K of T_C . In light of this, it is not surprising that we observed that reducing the field in the primary to 10 μ G did not change the data, except within about 0.1 K of T_C , it only reduced the signal-to-noise ratio.

RESULTS AND DISCUSSION

We have studied ten high-quality, nominally fully oxygenated YBCO films grown by pulsed-laser deposition, RF sputtering, and coevaporation with postannealing. Film thicknesses range from 500 to 2500 Å. Films are deposited onto SrTiO₃ (100) substrates, which are either 1-cm square or 15 mm diameter, circular. $2\theta/\theta$ x-ray scans show similarly grown films to be highly *c*-axis oriented with no extraneous phases present. Each film-growth technique is expected to produce a different microstructure enabling us to conclude that our results are independent of microstructure for high-quality films. Films *F-I* were made by laser ablation; *A, C,* and *J* were made by sputtering; and films *B, D, E* were made by coevaporation with postannealing. Even with these various deposition techniques, we note that there could still remain some property, such as epitaxial strain and generally higher defect densities, associated with all film-growth techniques, which would distinguish them from crystals. As indicated by the width of the peak in $\sigma_1(T)$, the films have sharp transitions, indicative of good homogeneity, with T_C 's ranging from 88 to 91 K. A typical transition width is less than 0.5 K, and the narrowest transition is less than 0.1-K wide, as determined from the full width at half maximum of the peak in $\sigma_1(T)$.

In all films presented here, $\lambda^{-2}(T)$ is a smooth, monotonically decreasing function with some slight variations (Fig. 1). We note that two of the three coevaporated films display the T -linear behavior at low temperature that is usually associated with a *d*-wave order parameter and very little disorder. Because the film thickness is less than the penetration depth, the two-coil method probes through the entire film. As a result of variations in growth parameters during

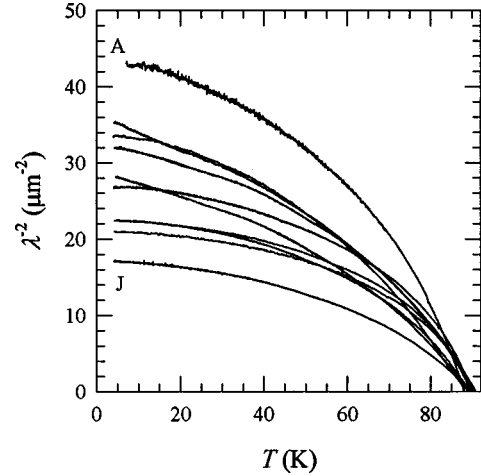


FIG. 1. $\lambda^{-2}(T)$ for the ten YBCO films studied. The films are labeled alphabetically in order of decreasing superfluid density at 4 K. Films *F-I* were made by laser ablation; films *A, C,* and *J* were made by sputtering; films *B, D,* and *E* were made by coevaporation with postannealing (films *G* and *H* overlap at low temperature).

deposition, the usual inhomogeneity associated with films is a variation in stoichiometry, hence in T_C , through the film thickness, as opposed to lateral variations. Whereas the resistivity of a film will drop to zero at the highest transition temperature, the mutual inductance shows all of the transitions as abrupt features in $\lambda^{-2}(T)$. None of the films presented here show signs of multiple transitions below 0.5 K of T_C . This, coupled with the highly oriented x-ray scans is indicative of high-quality films.

In the present paper we focus on $\lambda^{-2}(T)$ vs T near T_C . To make a general qualitative comparison of all of the films in the transition region, we normalize T to T_C and $\lambda^{-2}(T)$ to $\lambda^{-2}(0.96T_C)$, as is shown in Fig. 2. The behaviors of all of the films are in good agreement from $0.96 T_C$ to $0.99 T_C$. Although all of these samples are subject to defects inherent to films, the exceptional agreement over such a broad temperature range for films made by three different growth techniques leads us to believe that this behavior is representative of continuous films.

Above $0.99 T_C$, behavior is sample dependent. There are several plausible explanations for this behavior. To us, the most likely interpretation is that the films that show a drop in superfluid density do so as a result of a distribution of single-grain transition temperatures. (The reproducibility of the results at induced currents varying by an order of magnitude leads us to believe that the drop is not a result of Josephson-like coupling between the grains.) An upward curvature probably stems from slight variations in T_C through the film thickness due to slight variations in deposition conditions during film growth. To us, the most reasonable assessment of the data is that the films showing the sharpest transitions are those that are most representative of intrinsic behavior. The films with the sharpest transitions are those that most closely show mean-field ($2\beta=1$) behavior up to within 0.2 K of T_C . We include the wide variety of films primarily for completeness and to show that even what we believe to be less ‘‘intrinsic films’’ show mean-field-like behavior to within 0.5 K of T_C . Given that the variation in behavior from sample-to-sample is evident only in this region, we presume

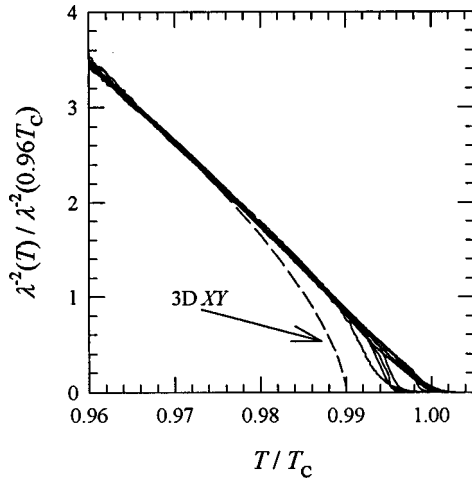


FIG. 2. $\lambda^{-2}(T)$ for all ten films normalized in temperature to T_C and in penetration depth to $\lambda^{-2}(0.96 T_C)$. All of the films show similar behavior in the range $0.96 T_C < T < 0.99 T_C$ and appear to be close to linear-in- T . Sample-to-sample variations become evident only within 1 K of T_C . The behavior expected of a 3D XY critical region, normalized at $0.96 T_C$, is shown as a dashed line.

that disorder effects do not play a significant role in determining behavior in the region $0.96 T_C < T < 0.99 T_C$.

Finally, we make a detailed quantitative analysis of each film. To determine the existence of a critical exponent we define the function

$$P(T) \equiv \frac{\lambda^{-2}(T)}{\partial \lambda^{-2} / \partial T}. \quad (8)$$

Both $\lambda^{-2}(T)$ and $\partial \lambda^{-2}(T) / \partial T$ are determined from the data. If $\lambda^{-2}(T) \propto (T_C - T)^{2\beta}$ over a temperature range of several Kelvins then we would observe,

$$P(T) = \frac{1}{2\beta} (T - T_C), \quad (9)$$

over that range. A linear least-squares fit to $P(T)$ vs T would yield both T_C and β . β should take the value $\beta = 1/3$ if 3D XY fluctuations dominate the physics, or some other value, identical from film to film, for other universal critical behaviors.⁹ Figure 3 shows $P(T)$ for three typical films, one made by each deposition technique. The line in Fig. 3 indicates 3D XY behavior, demonstrating that 3D XY is outside of experimental uncertainty. Least-squares fits over the temperature range from $0.96 T_C$ to about $0.995 T_C$ give values of 2β ranging from 0.80 to 1.20 for all of the films, including those not shown. In the sample with the sharpest transition, there is no significant deviation from MF behavior $2\beta = 1$ to within 0.2 K of a vanishing superfluid density. Since MF behavior is observed in all films to within 0.5 K of T_C , we set 0.5 K as an experimental upper bound on the width of the critical region in YBCO films. The film with the narrowest transition suggests that the intrinsic critical region may be smaller than 0.2 K. There does not exist any monotonic correlation between the magnitude of $\lambda(0)$ and the measured exponent, leading us to believe that the range of values found

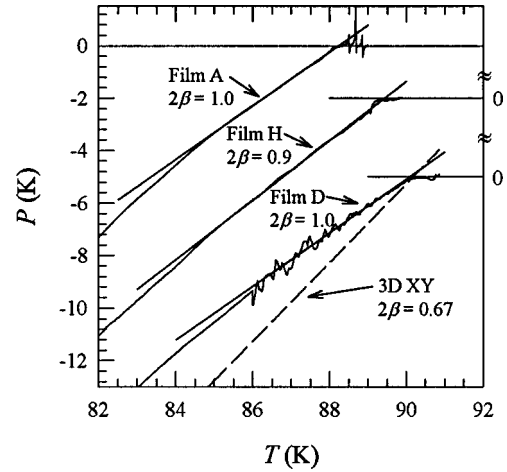


FIG. 3. $P(T) \equiv (1/\lambda^2) / [\partial(1/\lambda^2) / \partial T]$ is shown for a representative sputtered film A, a laser ablated film H, and coevaporated film D. Linear least-squares fits yielding the exponents $2\beta = 1.0, 0.9,$ and 1.0 , respectively, are shown as solid lines. The dashed line represents the linear behavior expected of a 3D XY exponent $2\beta = 0.67$. The data are offset along the ordinate for clarity.

for the exponent are simply variations in ‘‘mean-field’’ behavior associated with slight variations in film parameters and microstructure.

Let us consider how properties inherent to films may or may not affect the observed results with respect to the 3D XY behavior reported in crystals. In general, we expect defects inherent to films, such as grain boundaries, to enhance fluctuations. In a film where grain boundaries dominate the sheet inductance, we expect an even more dramatic suppression in the superfluid density than the $\beta = 1/3$ behavior of 3D XY. Microscopic defects such as dislocations or twins would nucleate extra fluctuations not found in crystalline samples. Moreover, given the fact that some of our films have $\lambda(0)$ significantly larger than the 1400 Å value associated with clean crystals, the Ginzburg criterion leads us to expect fluctuations to be stronger in films. The recent paper of Srikanth *et al.*⁸ confirms that high-purity crystals can exhibit a narrow critical range as well. It is plausible that the rapid decrease in quasiparticle scattering rate just below T_C observed in microwave cavity measurements on crystals²⁵ might impress an extra temperature dependence on the superfluid density since the superfluid density should be sensitive to the quasiparticle mean free path. Such a rapid decrease is not observed in films, and its absence is ascribed to the effects of weak disorder on a d -wave superconducting order parameter.

We return now to the issue of keeping measurements of $\lambda(T)$ within the linear-response regime. For illustration, two sets of $\lambda^{-2}(T)$ vs T data were taken on the same film, one set with a small current in the primary coil and the other with a much larger current, (inset to Figure 4). The data taken at low current are in the linear response regime, and they are linear in T to within 0.2 K of the transition. Data taken at higher current are in the nonlinear response regime for $T > 89$ K. The data are not shown, but coincident with the abrupt downturn in $\lambda^{-2}(T)$ at 89 K is an onset of a dissipative signal. Both features are expected if the nonlinearity comes from the generation of vortices by the field of the primary coil.

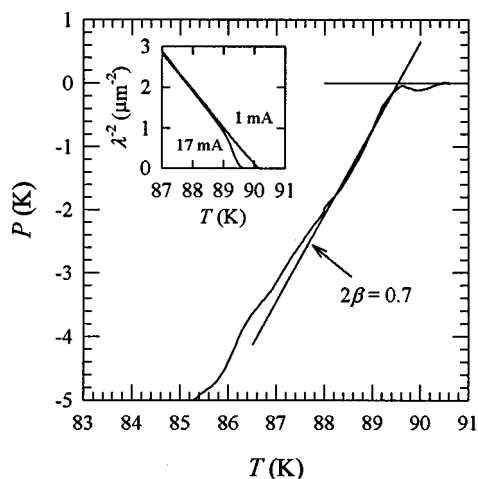


FIG. 4. $P(T)$ for film D taken in the nonlinear response range. The superfluid density is artificially suppressed, increasing the curvature in $\lambda^{-2}(T)$. A linear least-squares fit yielding an exponent $2\beta=0.7$, close to the expected 3D XY result, is shown as a solid line. The inset shows $\lambda^{-2}(T)$ measured both in the linear, drive-coil current=1 mA, and nonlinear, drive-coil current=17 mA, response regimes.

In the nonlinear regime, $\lambda^{-2}(T)$ vs T is reminiscent of the shape expected from 3D XY fluctuations. To quantify, we fit $P(T)$ from 88.0 to 89.4 K, the region that appears to the eye to behave as 3D XY. Figure 4 shows $P(T)$ for the high current data and the best fit, which yields $2\beta=0.70$, quite close to the 3D XY value $2\beta=0.67$. This result would lead us to erroneously identify a critical region 1.4-K wide, far larger

than the actual critical region of less than 0.2 K for this film. We emphasize again that great care has been taken to ensure that our measurements were performed in the linear-response regime to within 0.1 K of T_C .

CONCLUSION

There is no evidence for a large critical region in YBCO films from the magnetic penetration depth measured in samples made by sputtering, coevaporation, and pulsed-laser deposition. The critical region is less than 0.5 K wide in all of our YBCO films, and less than 0.2 K in the film with the sharpest transition. This upper bound on the range of critical behavior is consistent with the Ginzburg criterion. A small critical region indicates that fluctuations do not play as significant a role in YBCO as has been conjectured. It remains to be determined how their influence grows as $\lambda(0)$ is increased through oxygen depletion, chemical doping, or radiation damage, or as the film thickness decreases to single-unit-cell dimensions.

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