Crossover in temperature dependence of penetration depth $\lambda(T)$ in superconducting $YBa_2Cu_3O_{7-\delta}$ films

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(Received 25 June 1993)

The *ab*-plane penetration depth $\lambda(T)$ in twinned $YBa_2Cu_3O_{7-\delta}$ films exhibits a crossover in its T dependence at 25 K. Below 25 K, $\delta\lambda \equiv \lambda(T) - \lambda(0) \propto T^2$. This differs from recent measurements on a crystal which find $\delta\lambda \propto T$ below 40 K. Above 25 K, the normalized data $\lambda(T/T_c)/\lambda(0)$ vs T/T_c for our films agree extremely well with the crystal data, including quasilinear behavior between 25 and 40 K. Differences and similarities of films and crystals agree well with a theory of slightly disordered *d*-wave superconductors.

The average magnetic penetration depth $\lambda(T)$ of the *ab* plane of twinned $YBa_2Cu_3O_{7-\delta}$ samples has been studied often, but many issues remain unresolved. There is general agreement on the value $\lambda(0) = 1450$ Å ± 150 Å for the *ab* plane of fully oxygenated $YBa_2Cu_3O_{7-\delta}$, from muon spin rotation measurements on bulk samples.[1–4] Until recently, a consensus was developing that $\lambda^{-2}(T)/\lambda^{-2}(0) - 1$ at $T \ll T_c$ was close to T^2 for pure $YBa_2Cu_3O_{7-\delta}$, based primarily upon studies of films.[5–11] There was no agreement about the magnitude of the T^2 term,[7,11] however, or about the T dependence of λ over the entire range from 4 K to T_c. Indeed, most clean films[5–8,10] show $\lambda^{-2}(T)/\lambda^{-2}(0) \approx 1 - (T/T_c)^2$ over this range, but some[9,11,12] (all on substrates other than $SrTiO_3$) show an inflection point in $\lambda^{-2}(T)/\lambda^{-2}(0)$. Annett, Goldenfeld, and Renn[7] emphasized that the T^2 behavior could result from weak disorder in a $d_{x^2-y^2}$ superconductor, which would have T-linear behavior in the absence of disorder.

Into this unsettled situation came recently a report that $\lambda^{-2}(T)/\lambda^{-2}(0) - 1 \propto T$ from 4 to 40 K in fully oxygenated twinned $YBa_2Cu_3O_{7-\delta}$ crystals,[13] thus giving new impetus to the *d*-wave conjecture of Annett, Goldenfeld, and Renn[7] and motivating the present work. The theoretical sensitivity of *d*-wave superconductors to small amounts of disorder[14–17] immediately suggests this as an explanation of at least some of the reported sample-to-sample variations in λ for nominally identical pure $YBa_2Cu_3O_{7-\delta}$ samples. Of course, some variation may occur for other reasons, e.g., because macroscopic defects like grain boundaries might affect films more than crystals. In this paper, we show that our new data and much of the apparently conflicting data in the literature on $YBa_2Cu_3O_{7-\delta}$ films and crystals can be unified within the context of *d*-wave superconductivity. Moreover, in demonstrating this agreement, we show that the inductive response of films is primarily that of the grains and not the grain boundaries.

We present here measurements of $\lambda(T)$ from 4 K to T_c for three fully oxygenated $YBa_2Cu_3O_{7-\delta}$ films. We show that the data are in excellent agreement from 4 K to T_c with earlier, less precise, measurements made with a similar inductive technique on films made by a different process. We show that when film and crystal data are normalized as indicated by *d*-wave theory, they are in excellent agreement with each other above a crossover temperature of $T^* \approx 25$ K. We show that the qualitative and quantitative differences between films and crystals below the 25 K crossover are quantitatively explained if superconductivity is *d* wave and there is a slight residual disorder in the films that is absent in the crystals.

Our three films, *A*, *B*, and *C* are made by laser ablation of a stoichiometric sintered target onto (100) $SrTiO_3$ substrates, and they have their *c* axes perpendicular to the substrate. They are circular with diameters of 2.2 cm for films *A* and *C* and 2.0 cm for film *B*. All three film thicknesses are $d = 330 \pm 30$ Å. Superconducting transitions are 1 K wide near 88 K.

We determine $\lambda(T)$ from the mutual inductance $M(T)$ of concentric solenoidal coils on opposite sides of, and concentric with, each circular sample film, similar to Fiory *et al.*[5] and Jeanneret *et al.*,[18] but with solenoidal rather than counterwound coils. The primary coil comprises 84 turns of Cu wire; it is 3 mm long with inner and outer diameters of 3 and 7 mm. The secondary coil has the same dimensions as the primary coil, but comprises 300 turns of finer Cu wire. The film and substrate slip into a 2 mm gap between the coils. The inductive and dissipative parts of M are the voltages in the secondary circuit, divided by $I\omega$, at $\phi = 0°$ and $\phi = 90°$ relative to the sinusoidal current I at angular frequency ω in the primary circuit.

We checked that M was independent of the amplitude I and frequency of the current in the primary coil. Most data were taken at 20 kHz; data at 10 and 30 kHz were identical. M was independent of I when I was varied such that the maximum supercurrent density induced in the film ranged from 0.004 to 0.04 MA/cm². Most data

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were taken at the larger current. The magnetic field at the larger current was less than 1 mG perpendicular to the film, even at the film edges, and less than 0.2 G parallel to the film, so one would not expect the field to induce vortices. At 4.2 K, dissipation appears when the induced current density exceeds 10^6 and 10^7 A/cm^2 for films A and B, respectively. Based on a subsequent oxygen-depletion study of these films,[19] in which a small (< 1 mm diam) spot of film A detached from the substrate after annealing at 300 °C in Ar, we believe that the lower critical current density of A was from this defective spot, and not symptomatic of the entire film. The close agreement between films A and B suggests that the defective spot is negligible at the low experiment current densities.

The numerical model[6,10,18] relates the magnitude and phase of $M(T)$ to the conductivity $\sigma(T)=\sigma_1(T)+i\sigma_2(T)$ of the film by solving Maxwell's equations for the vector potential at each loop of the secondary coil due to the current in the primary coil and the induced currents in the film. The cylindrical symmetry of the apparatus simplifies the analysis considerably. The induced current density in the film is largest where the film is closest to the primary coil. It decreases rapidly toward the film edge, then just at the edge there is a much smaller peak. More than 2 K below T_c, $\sigma(T)=i\sigma_2(T)$ is purely inductive in our films, and we define $\lambda^{-2}\equiv\mu_0\omega\sigma_2$ ($\mu_0=4\pi\times10^{-7}$ H/m). Finally, the model allows for stray coupling between the primary and secondary circuits. For reference, the inductive and dissipative parts of $M(T)$ for film A are shown in Fig. 1. Note the sharp transition and the very rapid and large attenuation of M by the superconducting film within a few K below T_c.

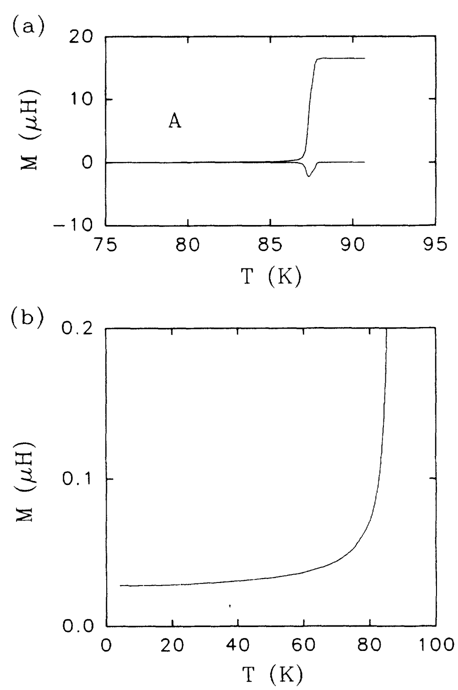

FIG. 1. The mutual inductance of the primary and secondary coils, M vs T, with film A inserted between them. (a) Inductive (upper) and dissipative (lower) components of M near T_c. (b) Magnified view of the inductive component of M at lower temperatures. The resistive component of M is zero.

While we use the full numerical model to obtain λ from M, for purposes of this discussion we introduce an approximate relationship between λ and M, which is accurate to better than 1% for $T<0.97T_c$. The approximation consists of taking the film diameter to be infinite, which is accurate because the film diameter is several times larger than the dimensions of the coils. In this approximation,[3,14]

$$M(T)=\alpha\lambda(T)\coth[d/\lambda(T)]+\beta\ . \qquad (1)$$

The geometric constant α is calculated to ±5% from the numerical model of the experimental apparatus. The constant β includes stray coupling between the primary and secondary circuits. The hyperbolic cotangent accounts for the decay of supercurrent density through the film thickness.[7] Our films are thin, $d \ll \lambda$, so one can further approximate: $M=\alpha\lambda^2/d+\beta$. The experimental uncertainty in β leads to an uncertainty in the value of λ(4 K) that we deduce from M(4 K). Hence, λ(4 K) is essentially a fitting parameter.

While the measured signal is proportional to λ^{+2}, the important microscopic quantity is the superfluid density, $\rho_s(T)\propto\lambda^{-2}(T)$, so the analysis will focus on λ^{-2}. It is useful to note that if $\lambda^2(T)-\lambda^2(0)\propto T^2$, then $\lambda^{-2}(T)-\lambda^{-2}(0)\propto T^2$ and $\delta\lambda\equiv\lambda(T)-\lambda(0)\propto T^2$, i.e., all such quantities have the same dependence on T. Plots of $\lambda^{-2}(T)$ or our three films are shown in Fig. 2, along with data on a twinned crystal.[13] Table I lists the parameters that characterize data.

There are three important points to be made here in connection with the uncertainty in β. First, determination of λ(4 K) to, say, ±10% from M(4 K)$-\beta$ demands a determination of β to ±20% of M(4 K). In our case, $M(T_c)\approx17\,000$ nH and M(4 K)$\approx$17 nH, so that $\beta\approx200$ nH must be measured to ±3 nH. Our actual uncertainty of ±30 nH locates $\lambda(0)$ between 0 and 3000 Å. Therefore, we fix β, hence λ(4 K)$\approx\lambda(0)$, by the criterion that our data agree with crystal data, as described below.

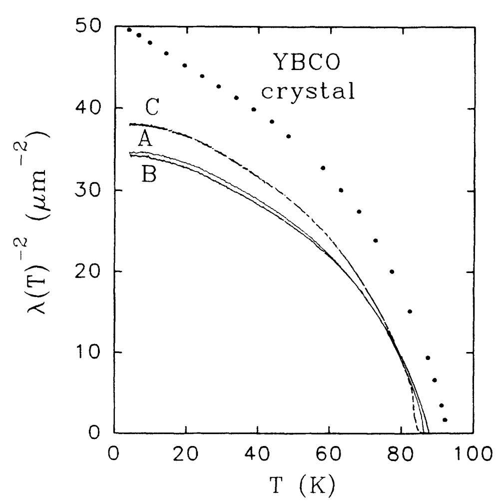

FIG. 2. $\lambda^{-2}(T)$ vs T for a $YBa_2Cu_3O_{7-\delta}$ crystal (Ref. 3) (solid circles) and for films A, B, and C.

TABLE I. T_c, $\lambda(0)$, c_0, and $1/c_2$ for $YBa_2Cu_3O_{7-\delta}$ films A, B, and C. $\lambda(0)$ is the penetration depth for the ab plane, $2c_2$ is the curvature of $\lambda^{-2}(T/T_c)/\lambda^{-2}(0)$ as $T/T_c \to 0$; c_0 is the fractional reduction in $\lambda^{-2}(0)$ in the films relative to $\lambda^{-2}(0)=(1400\ \text{Å})^{-2}$ in the crystal of Ref. 13.

Sample	T_c (K)	$\lambda(0)$	c_0	$1/c_2$ ($\pm 5\%$)
A	87.5	1700±50 Å	0.32	1.07
B	88.3	1710±50 Å	0.33	0.95
C	86.7	1630±50 Å	0.26	1.02

Second, despite the uncertainty in β, the value of $\lambda^{-2}(T)$ near T_c is known unambiguously because as T approaches T_c, $M \propto \lambda^2$ diverges and the uncertainty in β is negligible. Hence, our observation that λ^{-2} near T_c is only about 20% smaller in our films than in the crystal of Ref. 13 is certain, and it places a stringent upper limit of 20% on the possible contribution of grain boundaries to the total inductive response of the films. The third point is to make it clear that, in essence, we determine $\lambda^2(T)-\lambda^2(4\ \text{K})$ with considerable accuracy from $(d/\alpha)[M(T)-M(4\ \text{K})]$ because β is removed by the subtraction. Thus, the T^2 behavior that we observe, $\lambda^{-2}(T)/\lambda^{-2}(4\ \text{K})-1=c_2(T/T_c)^2$, is unambiguous. Moreover, since reasonable values for $\lambda(4\ \text{K})$ [$\approx\lambda(0)$] are not more than a few hundred angstroms above 1400 Å, as appropriate for reasonably clean material, the size c_2 of the quadratic term is known to roughly $\pm 20\%$, regardless of the uncertainty in β. Thus, the main points of this paper are insensitive to the uncertainty in β.

There are two sources of uncertainty. First, the $\pm 10\%$ uncertainty in the film thickness d and $\pm 5\%$ uncertainty in α translate into a $\pm 11\%$ uncertainty in the overall magnitude of λ^2, but not in its dependence on T. Second, the uncertainty in β affects the experimental value of $\lambda(4\ \text{K})$, as discussed above, and therefore the dependence of λ on T at low temperatures. In the following analysis, although β is the physical fitting parameter, we quote only the value of $\lambda(4\ \text{K}) \approx \lambda(0)$ that results from a particular choice of β; $\lambda(0)$ is effectively our fitting parameter.

The first goal of the analysis is to show that there exists a choice of β within experimental uncertainty that brings our results into excellent agreement with those of Hardy *et al.*[13] for $YBa_2Cu_3O_{7-\delta}$ crystals in the context of d-wave superconductivity. Note that our attempt to unify different experimental results differs from previous studies which focused on bringing measurements into agreement with theoretical and phenomenological expressions. There are many reasons why λ deduced from microwave measurements[13] on crystals at 1 GHz might differ from λ deduced from inductance measurement on films at 20 kHz, e.g., a greater influence of grain boundaries in films. However, it is noteworthy that both techniques measure, essentially, the inductance σ_2 of the superconducting electrons because dissipation due to σ_1 is extremely small even at 1 GHz, and that films and the crystal of Ref. 13 are twinned so that both experiments measure averages of σ_2 in the ab plane. Therefore, the excellent agreement that we find between films and the crystal is *a posteriori* justification of our assumption that both experiments probe the same physics, namely, the intrinsic superfluid density $\rho_s(T)$ of the ab plane of $YBa_2Cu_3O_{7-\delta}$.

To be specific about the agreement, when β is chosen for film A such that $\lambda(0)=1700$ Å, then the normalized data, $\lambda^{-2}(T/T_c)/\lambda^{-2}(0)$, is identical to $1.08\lambda^{-2}(T/T_c)/\lambda^{-2}(0)$ for the crystal above 25 K (Fig. 3). $\lambda(0)$ for the crystal is taken to be[1-4] 1400 Å. [The microwave measurements determine $\lambda(T)-\lambda(0)$, and one must choose $\lambda(0)$ to obtain $\lambda^{-2}(T)$.] The extremely small difference, upper panel of Fig. 3, equals the uncertainty in digitizing and interpolating the crystal data. We believe that it is highly unlikely that the film and microwave measurements are probing different physics, and that the agreement is accidental. Films B and C exhibit similarly good agreement. Deduced film parameters T_c, $\lambda(0)$, and the curvature c_2 appear in Table I. We emphasize the excellent agreement between 25 and 40 K, where the crystal data are clearly linear in T. A close examination of the derivative, $d\lambda^{-2}/dT$, indicates quasilinear behavior in the films as well. A discussion of this point, the fitting procedure, and other details can be found in Ref. 10. Finally, given the fitted values of $\lambda(0)$ for each film, Fig. 2 shows the unnormalized data and demonstrates that there is good film-to-film reproducibility.

The unusual normalization needed to bring the film and crystal data into coincidence is predicted in the theory of d-wave superconductivity.[14] In theory, $\lambda^{-2}(T) \propto T$ at $T/T_c \ll 1$ reflects the linear dependence of the "clean-limit" $d_{x^2-y^2}$ density of states, $N_s(E)$, on energy E for $E \lesssim 0.3\Delta_0$. If the cross section for scattering from point disorder is near the unitarity limit, then disorder creates an "impurity band" in $N_s(E)$ centered at

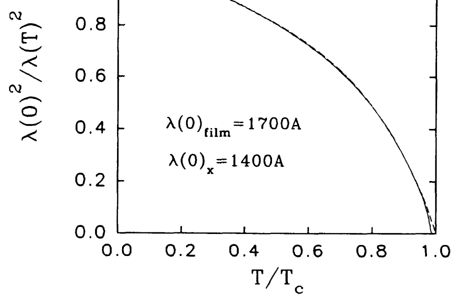

FIG. 3. (a) Difference between the curves in (b). The rms difference from 0.25 to $0.95T/T_c$ is 0.002. (b) Best fit above $0.25T_c$ of $\lambda^{-2}(T/T_c)/\lambda^{-2}(0)$ for $YBa_2Cu_3O_{7-\delta}$ film A and $1.08\lambda^{-2}(T/T_c)/\lambda^{-2}(0)$ for a crystal (Ref. 13). The crystal data are linear in T and lie above the film data as $T \to 0$.

$E=0$ by removing states from the logarithmic peak in N_s near Δ_0.[15] The number of states in the impurity band is proportional to the scattering rate Γ; the height $N_s(0)$ and width E^* of the impurity band are proportional to $\sqrt{\Gamma}$. At high temperatures, $kT>E^*$, the T dependence of λ^{-2} is largely determined by the T dependence of the order parameter $\Delta_0(T)$ and the high-energy structure in $N_s(E)$, and the impurity band serves only to reduce λ^{-2} slightly below its clean-limit value. At low temperatures, $kT<E^*$, most thermal excitations are in the impurity band, so the T dependence of λ^{-2} is altered dramatically from the clean limit. In other words, the qualitative prediction of theory is that clean and gently disordered materials should have the same T dependences above a crossover temperature when the experimental data for λ^{-2} are normalized to have the same slope near T_c, not when normalized to their respective values at $T=0$. The 8% difference in normalization in Fig. 3 serves this purpose.

This agreement is robust. We obtain similarly good agreement for any choice of $\lambda(0)$ for crystals in the accepted range from 1300 to 1700 Å. The corresponding "best fit" values of $\lambda(0)$ for films A and B range from 1630±50 Å to 1880±55 Å. Thus, our analysis finds that $\lambda(0)$ values for films A and B are not less than 15% or more than 30% larger than for clean crystals. Corroborating evidence is the observation that the values of $\lambda^{-2}(T)$ near T_c for our films are correspondingly smaller than for the crystal (see Fig. 2). On the other hand, when we fit to the commonly used function $\lambda^{-2}(T)/\lambda^{-2}(0)\approx 1-(T/T_c)^2$ from 4 K to nearly T_c, the best agreement is about ±2%, even when fitting only over a restricted range from 25 to 85 K.[10] This is not bad, but the fit to the crystal data is *ten times better* (upper panel of Fig. 3).

We have compared our films with published data[6] on two 500 Å and one 2000 Å thick films of $YBa_2Cu_3O_{7-\delta}$ made by codeposition and postannealing onto $SrTiO_3$ and $LaAlO_3$ substrates. Assuming that these authors actually determined $\lambda(T)\coth[d/\lambda(T)]-\lambda(0)\coth[d/\lambda(0)]$ and then obtained $\lambda(T)$ effectively by assuming a value for $\lambda(0)$, we can report their data for other choices of $\lambda(0)$. Their results agree with ours, within their noise which is several times larger than ours, when their values of $\lambda(0)$, obtained from a best-fit to BCS, are adjusted upward from 1500, 1660, and 2100 Å to 1770, 1910, and 2700 Å, respectively.[10] While the large value $\lambda(0)=2700$ Å suggests that disorder in this film is much larger than in the other films, the other two films clearly behave in all respects very much like our three films.

Having shown that several films from different laboratories agree with each other and with a crystal, when compared in the framework of d-wave superconductivity, we now turn to a quantitative analysis of the differences at low T. Hirschfeld and Goldenfeld[14] have calculated the effects of slight disorder on $\lambda^{-2}(0)$ and the curvature in $\lambda^{-2}(T)/\lambda^{-2}(0)-1$ at $T\ll T_c$. They neglect complications like dispersion along the c axis, localized states, a Van Hove singularity in the normal-state density of states, and the possible influence of disorder on normal-state parameters like the charge-carrier density and the spectrum of antiferromagnetic spin fluctuations. The theory is insensitive to its approximations when the penetration depth $\lambda(T)$ in the disordered material is normalized to the clean-limit penetration depth at $T=0$, λ_0.

Specifically, at $T/T_c\ll 1$ and small disorder, $\gamma\ll\Delta_0$ [$\gamma\approx 0.63(\Gamma\Delta_0)^{1/2}$], theory predicts that disorder decreases $\lambda^{-2}(0)$ and decreases the curvature in $\lambda^{-2}(T)$:

$$\frac{\lambda_0^2}{\lambda^2(T)}=1-\frac{2\gamma}{\pi\Delta_0}\ln\left[\frac{4\Delta_0}{\gamma}\right]-\frac{\pi}{3\Delta_0\gamma}T^2 =1-c_0-\frac{\pi}{3\Delta_0\gamma}T^2 \tag{2}$$

and

$$\frac{\lambda^2(0)}{\lambda^2(T/T_c)}=1-\frac{\pi T_c^2}{3\Delta_0\gamma(1-c_0)}\frac{T^2}{T_c^2} =1-c_2\frac{T^2}{T_c^2}\ . \tag{3}$$

Equation (2) defines c_0 to be the fractional change in $\lambda^{-2}(0)$ from disorder for fixed λ_0 (see Table I). Equation (3) defines $2c_2$ to be the curvature of $\lambda^2(T/T_c)/\lambda^2(0)$. Assuming that Δ_0/kT_c is a weak function of disorder, then theory finds $c_0=(2x/3)\ln(12/\pi x)$ and $c_2=T_c^2/[\Delta_0^2x(1-c_0)]$ are functions of the disorder parameter $x\equiv 3\gamma/\pi\Delta_0$, and can be plotted against each other, thereby removing the unmeasured x from the comparison.

The solid and dashed curves in Fig. 4 show $1/c_2$ vs c_0 calculated for $\Delta_0/kT_c=3$ and 3.5, respectively. This value of Δ_0/kT_c is supported by the observation that $\lambda^{-2}(T)/\lambda^{-2}(0)-1$ in the crystal of Ref. 13 is linear in T

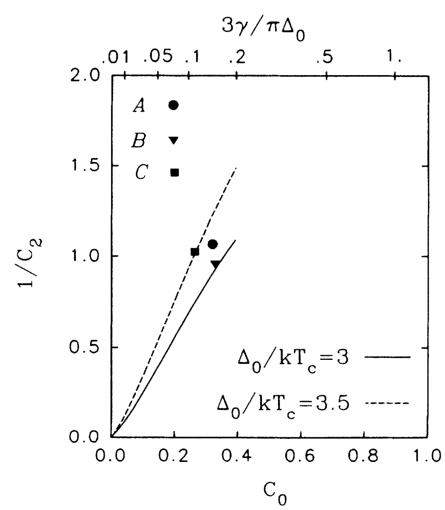

FIG. 4. The curves represent $1/c_2$ vs c_0 for $\Delta_0/kT_c=3$ and 3.5, from the $d_{x^2-y^2}$ theory of Hirschfeld and Goldenfeld (Ref. 14), which is valid for small disorder, $3\gamma/\pi\Delta_0\ll 1$, as marked on upper scale. Points represent our films; crystal data (Ref. 13) would be at $1/c_2=c_0=0$. $2c_2$ is the curvature of $\lambda^{-2}(T/T_c)/\lambda^{-2}(0)$ as $T/T_c\to 0$; c_0 is the fractional reduction in $\lambda^{-2}(0)$ in the films relative to $\lambda^{-2}(0)=(1400\ \text{Å})^{-2}$ in the crystal.

up to 40 K, which requires that $N_s(E)$ is linear in E up to about $k \times 100$ K, which requires that the logarithmic singularity in $N_s(E)$ occurs near $k \times 270$ K $\approx 3kT_c$.[20] The three data points from our films are in excellent agreement with the theory curves with $\gamma/\Delta_0 \approx 0.15$ (see top scale in Fig. 4). (The crystal data would be at $c_0 = 1/c_2 = 0$. The infinite curvature, $1/c_2 = 0$, for the crystal means that the low-T parabolic behavior is as sharp as the vertex of a triangle.) In other words, the low-T curvature in λ^{-2} corresponds quantitatively to the deduced suppression in superfluid density. Moreover, the scattering rate Γ found from γ is quite small, $\Gamma \approx 0.2kT_c/\hbar$, consistent with a small residual resistivity of about 20% of the resistivity just above T_c. Thus, the films behave at low T just as d-wave superconductivity theory predicts, assuming that the films have a slight residual disorder that is absent in the crystal of Ref. 13.

In conclusion, $\lambda(T)$ data in our laser-ablated films of $YBa_2Cu_3O_{7-\delta}$ agree with published data on films made differently. They quantitatively agree with data on a crystal[13] within the framework of a microscopic theory[14] of d-wave superconductivity, with the assumption that there is a slight residual disorder in the films which is absent in the crystal. The observation that the slope of λ^{-2} near T_c in the films is only about 20% lower than in crystals puts an upper limit of 20% on the possible contribution of grain boundary Josephson junctions in the films to their total sheet inductance. Therefore, our results establish that it is possible to make $YBa_2Cu_3O_{7-\delta}$ films of sufficient quality to study the intrinsic properties of the material. Reports[9,11,12] of T dependences for λ^{-2} greatly different from ours come from films on substrates other than $SrTiO_3$, which is generally acknowledged as the substrate which results in the highest quality films. Measurements on intentionally disordered, doped films follow the d-wave theory well, too, in that $\lambda^{-2}(0)$ decreases much more rapidly with doping than possible from conventional s-wave superconductivity.[20] Thus, in total, penetration depth measurements in films and crystals strongly support the idea that superconductivity in $YBa_2Cu_3O_{7-\delta}$ is d wave.

The authors acknowledge valuable discussions with Daniel Cox, Nigel Goldenfeld, Peter Hirschfeld, and Yong-Jihn Kim, computer programming by Ki Ho Lee, and help from Eric Ulm, Dimitrios Xenikos, and Jin-Tae Kim. The work at OSU was supported by DOE Grant No. DE-FG02-90ER45427 through the Midwest Superconductivity Consortium.

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