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Measurements of the temperature dependence of the magnetic penetration depth in $YBa_2Cu_3O_{7-\delta}$ superconducting thin films

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Using a very sensitive microstrip-resonator technique, we have measured $\lambda(T)$ of c-axis $YBa_2Cu_3O_{7-\delta}$ films on various substrates and compared the results to similar data on Nb and NbCN films. For $YBa_2Cu_3O_{7-\delta}$, $\lambda(T)$ is found to be exponential and not power law in its temperature dependence (for $T < T_c/2$) in the best films. Overall the data cannot be described by a single-gap BCS temperature dependence. The data at low temperature provide a lower limit of $2\Delta(0)/k_BT_c \approx 2.5$ for the minimum gap in $YBa_2Cu_3O_{7-\delta}$, yet show strong-coupling-like behavior at high temperatures. We discuss our results in the context of other experiments on the cuprate superconductors.

It is well known that the temperature dependence of the superconducting magnetic penetration depth $\lambda(T)$ is sensitive to the symmetry of the electron-pairing state leading to superconductivity and hence the presence of nodes in the pair wave function for certain directions in k space. Previous measurements of $\lambda(T)$ for the high-temperature copper oxide superconductors are most consistent with a BCS nodeless pair wave function.[1–3] On the other hand these experiments generally lose sensitivity in the low-temperature regime where the differences between the various predictions are most distinct. Also, recent infrared studies of untwinned single crystals of $YBa_2Cu_3O_{7-\delta}$ (Y-Ba-Cu-O) show a clear in-plane anisotropy along the a and b crystallographic directions,[4] which should lead to deviations from simple isotropic BCS behavior at all temperatures. And finally, there has been considerable controversy in the literature concerning the true temperature dependence of $\lambda(T)$ in Y-Ba-Cu-O (Ref. 5) and how to extract values of $\lambda(0)$ from the data. Thus measurements of $\lambda(T)$ remain central to our understanding of the high-temperature copper oxide superconductors.

In this paper we present a different, particularly sensitive approach to measurements of $\lambda(T)$. It is based on measurements of the surface inductance of a superconductor in thin-film form contained in a microstrip resonator.[6–8] The basic idea of the measurement is to deduce the penetration depth from a measurement of the phase velocity of a microwave signal propagating on a thin-film superconducting transmission line. In the particular case of a microstrip transmission line (see the inset of Fig. 1), the dependence is given by[9]

$$\frac{v_p(T)}{c} = \frac{1/(\epsilon_{\rm eff})^{1/2}}{\{1+2[\lambda(T)/d]\coth[t/\lambda(T)]\}^{1/2}}, \qquad (1)$$

where d is the dielectric thickness. $\epsilon_{\rm eff}$ is the effective relative dielectric constant, and both films in the transmission line are of thickness t and have penetration depth $\lambda(T)$. With this technique it is possible to measure changes in λ of less than 1 Å, an order of magnitude better than other techniques used to date.[8]

Two identically prepared c-axis Y-Ba-Cu-O superconducting films [thickness $t > 3\lambda(0)/2$] on separate substrates are used to make a microstrip transmission line resonator by sandwiching a thin dielectric sheet (e.g., Teflon FEP) between the films and clamping the whole assembly.[6–8] For our c-axis oriented films, the shielding currents for the propagating mode flow entirely in the direction parallel to the ab plane. However, since the Y-Ba-Cu-O films are heavily twinned, shielding currents flow roughly equally along the a- and b-crystallographic directions. The measurements are performed at excitation levels for which the data are independent of the applied microwave power.

An attractive feature of this measurement technique is that the data can be used directly to reveal the temperature dependence of $\lambda(T)$ at low temperatures, in a manner that is independent of the microstrip geometry (t, d, w, and $\epsilon_{\rm eff}$) and any assumed theoretical temperature dependence of $\lambda(T)$.[7,8,10] Rewriting Eq. (1), subtracting off a zero-temperature phase velocity baseline, gives in the thick- and thin-film limits,

$$\ln\left[\left(\frac{c}{v_p(T)}\right)^2 - \left(\frac{c}{v_p(0)}\right)^2\right] = \ln\left(\frac{\lambda(T)}{\lambda(0)} - 1\right) + \text{const.} \quad \begin{pmatrix} T < T_c/2, & t < \lambda/2 \\ T < T_c/2, & t > 3\lambda/2 \end{pmatrix}, \qquad (2)$$

where the constant is independent of temperature, except for a possible small temperature dependence of $\epsilon_{\rm eff}$ which has been shown to be absent in the data presented here.[8,11] For a local clean-limit BCS-like superconductor with a finite gap in all directions in k space, $[\lambda(T)/\lambda(0) - 1] \propto T^{-1/2}\exp[-\Delta(0)/k_BT]$ as T goes to zero,[12] whereas for a superconductor with nodes in the gap, $[\lambda(T)/\lambda(0) - 1] \propto (T/T_c)^n$ as T goes to zero, the value of n depending on the specific nature of the nodes (points, lines, etc.).[13]

The data typical of the best high-T_c superconducting Y-Ba-Cu-O films on MgO single-crystal substrates are plotted in Fig. 1 so as to test the exponential temperature dependence. A test of the power-law dependence is shown

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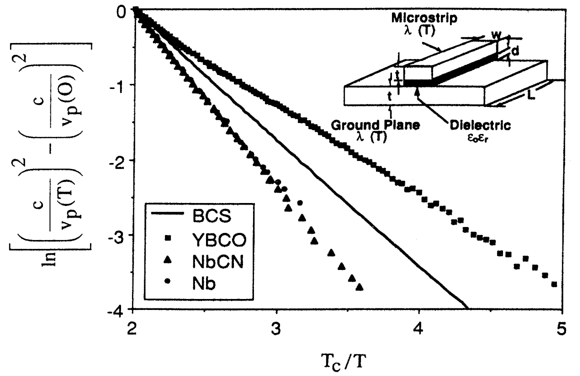

FIG. 1. Raw data in the form $\ln\{[c/v_p(T)]^2-[c/v_p(0)]^2\}$, which is proportional to $\ln[\lambda(T)/\lambda(0)-1]$, is plotted vs T_c/T to emphasize any exponential decay of $\lambda(T)$ for Nb, NbCN, and Y-Ba-Cu-O/MgO (sample M058C45) films. Also shown as a solid line is the temperature dependence $\ln[\lambda(T)/\lambda(0)-1]$ calculated for BCS weak-coupling theory (Ref. 12). The inset shows the geometry of the microstrip transmission line.

in Fig. 2. Only for $T < T_c/2$ are the slopes of these lines suitable for establishing the low-temperature limiting behavior,[14] and there the slopes can be interpreted as $-\Delta(0)/k_B T_c$ and n in Figs. 1 and 2, respectively. Comparable data for Nb and NbCN are also shown as a test of our technique.[15] In Fig. 1 the data for Nb, and NbCN films deposited on sapphire show straight lines for $T < T_c/2$ as expected for a BCS superconductor. From the slopes of the lines we find $2\Delta(0)/k_B T_c = 4.4 \pm 0.5$ and 4.8 ± 0.5 for Nb and NbCN, respectively, somewhat higher than expected.[14] The low-temperature slope observed for the Y-Ba-Cu-O/MgO film corresponds to $2\Delta(0)/k_B T_c = 2.5 \pm 0.3$ [or $2\Delta(0) = 17.5 \pm 2$ meV $= 140 \pm 16$ cm^{-1}]. From Fig. 2, an exponent n of about 3 is obtained.[8,16] Examination of Figs. 1 and 2 shows that our Y-Ba-Cu-O data are better described by an exponential temperature dependence than by a power law. While the exact shape of these curves is sensitive to the choice of a zero-temperature baseline, $v_p(0)/c$, the conclusions above regarding exponential versus power-law fits are not.[14] Observation of exponential behavior at low temperature does not absolutely rule out non-s-wave pairing.[13] Strictly speaking it only says that there are no nodes where the density of states is finite.

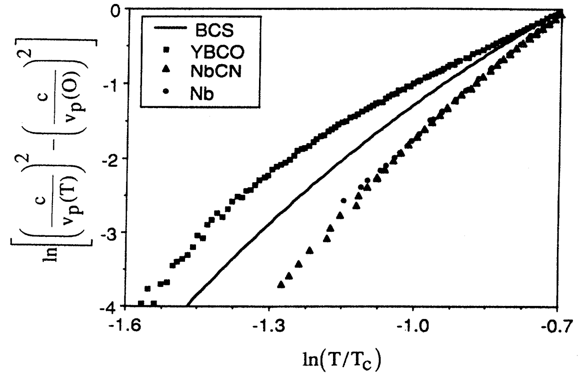

FIG. 2. The data from Fig. 1 are now plotted vs $\ln(T/T_c)$ to emphasize any power-law decay of $\lambda(T)$.

A possible origin of the low values of $2\Delta(0)/k_B T_c$ found for Y-Ba-Cu-O may be the nature and density of defects present in these films. We choose to concentrate on the Y-Ba-Cu-O/MgO films because it is known from transport studies that they have better normal-state properties than those on YSZ or $LaAlO_3$.[10,16] In addition, the best films on MgO studied here have a normal-state resistivity at 100 K of 60 $\mu\Omega$ cm,[10] an rf residual loss of 16 $\mu\Omega$ at 10 GHz, 4.2 K,[17] an absolute value of $\lambda(0) \approx 1400$ Å,[10] and a low density of high-angle grain boundaries,[17] making them among the best high-T_c films available. As already mentioned, like all films they are highly twinned. Nevertheless the transport properties of these Y-Ba-Cu-O/MgO films are comparable to single crystals and their rf properties are in fact as good or superior.

Up to this point, we have focused on only the temperature dependence of $\lambda(T)$ for $T < T_c/2$. To examine the data at higher temperatures, we must extract $\lambda(T)$ from the phase velocity data over the entire temperature range. As discussed in detail elsewhere,[6–8,11] this is done by self-consistently fitting the data to an assumed theoretical temperature dependence. The data for Nb and NbCN can be fit very well with a BCS temperature dependence over the entire temperature range of the measurement using a single energy gap[11,18] with values of $2\Delta(0)/k_B T_c \approx 4.2$ and 4.75, respectively (see Fig. 3). The values of the gaps are still higher than tunneling results, which give $2\Delta(0)/k_B T_c = 3.8$ for Nb,[19] however they are comparable to recent stripline surface resistance measurements which yield values of $2\Delta(0)/k_B T_c = 4.4$ for Nb, and 4.75 ± 0.25 for NbN.[20] The value of $\lambda(0)$ in the sputtered Nb film is found to be approximately 825 Å, in good agreement with previous measurements,[21] while $\lambda(0) = 2820$ Å in the NbCN film.

In the case of our Y-Ba-Cu-O/MgO films, we find that the temperature dependence of λ cannot be self-consistently fit to a BCS temperature dependence derived from a single energy gap over the entire temperature range.[22] From Fig. 1, it is clear that the low-temperature data can be fit to a BCS temperature dependence with a single gap of $2\Delta(0)/k_B T_c = 2.5 \pm 0.3$.[7] By examining the temperature dependence of $[\lambda(0)/\lambda(T)]^2$, we find that the high-temperature ($0.65T_c$ to $0.98T_c$) data best fits a single-gap BCS temperature dependence with an effective $2\Delta(0)/k_B T_c = 4.5 \pm 0.25$. This is in good agreement with measurements of $\lambda(T)$ restricted to this same temperature range in single-crystal Y-Ba-Cu-O (Ref. 23) where it was found that a single-gap BCS fit yields $2\Delta(0)/k_B T_c = 4.3$. Thus it seems that the electrodynamic response of the cuprate superconductors provides additional evidence for large $2\Delta(0)/k_B T_c$ in these materials, although lacking any direct evidence for the validity of isotropic BCS theory, it is not clear how accurate the inferred values of $2\Delta(0)/k_B T_c$ actually are.

Based on our data, we conclude that the temperature dependence of $\lambda(T)$ at low temperatures is exponential and not power law, and the size of the parameter $2\Delta(0)/k_B T_c$ is at least 2.5 for Y-Ba-Cu-O. The penetration depth data at high temperatures shows strong-coupled-like behavior, in sharp contrast to the low-temperature behavior. We now discuss these results more broadly in the context of what is presently understood about the cuprate superconductors.

Early work on superconductor electrodynamics was based on an empirical two-fluid model, in which a normal and superconducting electronic fluid coexist in the material.[24] A two-fluid model consistent with the residual surface resistance of our Y-Ba-Cu-O/MgO films,[17,25] leads to the conclusion that roughly 25% of the charge carriers remain unpaired in Y-Ba-Cu-O at low temperatures.[17] However, this model is inconsistent with our observations of exponential behavior in $\lambda(T)$ at low temperatures.

A variation on the two-fluid model assumes that Y-Ba-Cu-O consists of two shielding superfluids, the planes and the chains, which act in parallel,[26] as possibly suggested by recent infrared data on untwinned single crystals.[4] This model is consistent with measurements of two distinct nuclear-spin relaxation rates $1/T_1T$ in Y-Ba-Cu-O,[26] and recent Raman scattering results from $Y_2Ba_4Cu_8O_{16-x}$ suggesting two gaps $[2\Delta(0)/k_BT_c=6.3,2.8]$.[27]

The two fluids, in general, will have different oscillator strengths n_s/m^* in the superfluid response, so the composite penetration depth will be of the form $\lambda_{\text{parallel}}^{-2}=\mu_0e^2(n_{s1}/m_1^*+n_{s2}/m_2^*)$. Presumably each component has its own energy gap $2\Delta_{1,2}(0)/k_BT_c$. For the Y-Ba-Cu-O film on MgO shown in Fig. 1, the observed temperature dependence can be self-consistently fit (with 3 parameters) within this model using a choice of two BCS temperature dependencies with energy gaps $2\Delta_1(0)/k_BT_c=2.0$ and $2\Delta_2(0)/k_BT_c=4.5$, and an oscillator strength weighting factor $(m_1^*/n_{s1})/(m_2^*/n_{s2})\approx 3.5$ (see the Y-Ba-Cu-O/MgO curve in Fig. 3). The zero-temperature penetration depth resulting from this fit is $\lambda_{\text{parallel}}(0)=1565$ Å, which yields $\lambda_1(0)=3320$ Å and $\lambda_2(0)=1775$ Å for the two components.

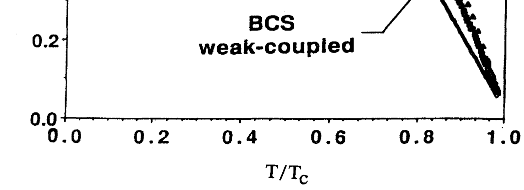

FIG. 3. Here $[\lambda(0)/\lambda(T)]^2$ is plotted vs T/T_c for self-consistent fits of the Nb (a strong-coupling superconductor) and Y-Ba-Cu-O/MgO films. The Y-Ba-Cu-O data were self-consistently fit to the models discussed in the text, with results that are indistinguishable on this plot. The curve calculated for BCS weak-coupling theory (Ref. 12) is also shown.

However, from estimates of the anisotropy of the plasma frequency measured by ir reflectivity in untwinned Y-Ba-Cu-O single crystals by Schlesinger *et al.*,[4] one infers an oscillator strength ratio (in the clean limit) in the *ab* plane of $(m_{\text{chains}}^*/n_{\text{chains}})/(m_{\text{planes}}^*/n_{\text{planes}})\approx 0.5$, very different from that found in the parallel superconductor fit to our data. Thus, the measured penetration depth and surface resistance[17] data at microwave frequencies are not consistent with published ir reflectivity measurements of Y-Ba-Cu-O within a parallel superconductor model. In addition, this model does not explain the origin of the residual microwave surface resistance seen in these same Y-Ba-Cu-O/MgO films.

A third interpretation of our data is that weak links at twin boundaries are influencing the electromagnetic response. Weak-link structures (caused for example by oxygen vacancy-ordered defects) may also be present even in untwinned single crystals.[28] The combined effects of grains and weak links can be modeled as a network of inductors, yielding an effective penetration depth, in the small grain limit ($a\ll\lambda$) of[29]

$$\lambda_{\text{series}}(T)=\{\lambda_{\text{SC}}^2(T)+\Phi_0/[2\pi\mu_0aJ_c(T)]\}^{1/2},$$

where $\lambda_{\text{SC}}(T)$ is the penetration depth of a perfect grain and a is the weak-link spacing. For our samples reasonable estimates for the fixed parameters of this model are a BCS penetration depth with $T_c=90$ K and $\lambda_{\text{SC}}(0)=1400$ Å for the domains,[22] with twins spaced about 0.1 μm apart (as seen in TEM micrographs[16]) film thickness of 4340 Å and an interdomain $J_c(0)=2\times10^7$ A/cm^2.[17,30] For the weak-link critical current temperature dependence, we take the Ambegaokar-Baratoff form,[31] in order to be consistent with our observations of an exponential dependence of $\lambda(T)/\lambda(0)-1$ at low temperatures. Two parameters were varied to get a best fit to the data: $2\Delta(0)/k_BT_c$ in the domains [which strongly influences the high-temperature dependence of $\lambda(T)$] and $\Delta^*(0)$, the reduced gap at the weak link (which influences the low-temperature fit). We obtain excellent self-consistent fits to the data (indistinguishable from that shown in Fig. 3) on a film with a small large-angle grain boundary content by taking a $2\Delta(0)/k_BT_c=4.5$ and a weak link $\Delta^*(0)$ of about 6-7 mV [giving an $I_cR_{NN}=10.5$ mV, comparable to that consistent with the 10 GHz residual losses on the same film[17] and also consistent with a minimum gap in Y-Ba-Cu-O of $2\Delta(0)/k_BT_c\geq 2.5$]. The zero-temperature penetration depth for this fit is $\lambda_{\text{series}}(0)=1395$ Å.[32,33] Direct measurement of the weak-link parameters of twin boundaries will be necessary to prove or disprove their role in the microwave properties of the cuprate superconductors.[34] However, one can use the parameters for this model and calculate the far-infrared absorption in the films. These calculations give an absorptivity of approximately 1% between 100 and 400 cm^{-1}, in good agreement with recent infrared measurements on these films,[35] and single crystals.[36]

In summary, we have examined the temperature dependence of the magnetic penetration depth of a variety of Y-Ba-Cu-O thin films. The low-temperature dependence is exponential, arguing against the possibility of nodes in the energy gap in Y-Ba-Cu-O (at least for directions with finite density of states). We have measured the minimum energy gap from the low-temperature dependence of $\lambda(T)$ of $2\Delta(0)/k_BT_c \gtrsim 2.5$. Unlike conventional strong-coupled superconductors, the inferred temperature dependence of $\lambda(T)$ in the Y-Ba-Cu-O/MgO films is not characterized by a single-gap (scaled, isotropic) BCS temperature dependence. The physical origin of this behavior remains an open question.

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[14] The slopes of the lines in Figs. 1 and 2, as well as their shape, are somewhat sensitive to the choice of $v_p(0)/c$, which comes from extrapolation of the data to $T=0$. We are more confident about the slopes derived from the Y-Ba-Cu-O data because it extends down to $T/T_c=0.1$.

[15] The Nb and NbCN films were sputtered by J. Murduck at TRW, Redondo Beach, from Nb targets in Ar and Ar-N_2-CH_4 atmospheres, respectively. The Nb film has $T_c=9.2$ K and residual resistivity ratio (RRR) of 4 with $\rho(0)=4\ \mu\Omega$ cm and thickness 3650 Å. The NbCN film has a $T_c=15.8$ K, RRR of 1.1, $\rho(0)=60\ \mu\Omega$ cm, and thickness 5660 Å.

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[32] The fit values of $\lambda_{\rm series}(0)$ is close to the value of 1400 Å taken for the domains, but is quite different from the value calculated from the equation for $\lambda_{\rm series}(T)$. This is because fits to the data do not make use of the magnitude of the fitting function, only its temperature dependence.

[33] In general we find a strong dependence of $\lambda(0)$ on the temperature dependence used to fit the data (see Refs. 6–8, 11) as others have (see Ref. 5). This problem is common to all measurement techniques that do not uniquely determine a zero-temperature baseline.

[34] If the weak links are taken to be grain boundaries [with $a=1$ μm (Ref. 16) and $J_c(0)=2\times10^6$ A/cm^2] instead of the twin boundaries, the fit parameters are almost identical: $2\Delta(0)/k_BT_c=4.5$, $\Delta^*(0)=8$ meV. Hence the weak links can originate from several different kinds of defects.

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