RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 40, NUMBER 10 1 OCTOBER 1989

Millimeter-wave surface impedance of $YBa_2Cu_3O_7$ thin films

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(Received 12 June 1989)

We have measured the millimeter wave ($f=101.3$ GHz) surface impedance of various $YBa_2Cu_3O_7$ films deposited on various substrates. $R_s(T)$ rapidly decreases below T_c and is, at $T=77$ K, orders of magnitude smaller than that found generally in ceramic and thin-film specimens prepared by other groups. A comparison is given with predictions of the two-fluid model and the BCS theory.

The electromagnetic response of various high-temperature superconductors has been thoroughly explored using a variety of experimental techniques. Early measurements[1] of the penetration depth $\lambda(T)$ have indicated deviations from the BCS behavior, while recent experiments[2] performed on single crystals are in agreement with conventional pairing. Optical experiments[3] are controversial, and gap values both in agreement with weak-coupling BCS theory and far exceeding this limit have been frequently reported in the literature.

In this Rapid Communication we present experimental results and analysis of the surface impedance Z_s. When $\xi_0 \ll \lambda$, as it is for the oxide superconductors, the surface impedance is related to the complex conductivity $\sigma=\sigma_1-j\sigma_2$ by

$$Z_s=\left(\frac{j\mu_0\omega}{\sigma_1-j\sigma_2}\right)^{1/2}=R_s+jX_s\,. \tag{1}$$

Here μ_0 is the permeability of free space and $\omega/2\pi$ the measurement frequency. R_s and X_s are the surface resistance and the surface reactance, respectively.[4]

The surface resistance $R_s(T)$ has been measured by various groups on both ceramic, thin-film, and single-crystal specimens.[4] Ceramic and thin-film materials usually have substantial residual surface resistance $R_s(T=0)$, and $R_s(T)$ also displays drastic deviations from the BCS behavior [i.e., linear temperature dependence of $R_s(T)$ below 50 K]. The residual losses are most probably due to imperfections, grain boundaries, etc., which can be modeled[5] in terms of a network of Josephson junctions. The power-law temperature dependence[6] of $R_s(T)$ and X_s, however, have not been accounted for. Experiments on high-quality single crystals[7] and oriented thin films[8] show a significant reduction of $R_s(T)$ in the superconducting state where the experiments are limited to the immediate vicinity of T_c due to a lack of sensitivity associated with the relatively low measurement frequency. Because of this, and also because of complications arising from the finite thickness of the films, a detailed comparison with calculations has not been performed.

Our experiments have been conducted on $YBa_2Cu_3O_7$ films deposited on $SrTiO_3$ and $LaAlO_3$ substrates by means of a pulsed-laser deposition process described elsewhere.[9,10] The substrate surface during deposition was kept at temperatures between 650 and 750 °C with a background oxygen pressure of 220 mTorr. These *in situ* deposited films, without post annealing, exhibited zero resistance transition temperatures of over 91 K with onset at 93 K and dc critical current densities of over 5×10^6 A/cm^2 at 77 K. The films have a crystalline structure close to that of bulk single crystals as measured by an ion channeling technique,[11] showing a minimum yield of 3%. This represents nearly perfect *c*-axis alignment.

The surface resistance $R_s(T)$ and surface reactance $X_s(T)$ has been measured in a cylindrical copper TE_{011} transmission cavity operating at 101.3 GHz. In this configuration, currents flow in the Cu-O planes, and consequently the measured R_s and X_s are related to the in-plane properties. We measure the resonance frequency and bandwidth as a function of temperature using the superconducting film or a polished copper plate as the bottom of the cavity. The difference between the sample surface impedance and the copper surface impedance is proportional to the change in the bandwidth $\Delta W=W_{\text{sample}}-W_{\text{Cu}}$ and the resonance frequency $\Delta f=f_{\text{sample}}-f_{\text{Cu}}$:

$$\Delta Z_s=\Delta R_s+j\Delta X_s=\gamma^{-1}(\Delta W/2-j\Delta f)\,, \tag{2}$$

where γ is the resonator constant for the mode. With R_s (copper) known, R_s ($YBa_2Cu_3O_7$) can be evaluated. Our experiments on the reactance X_s will be reported later. Here we describe our experiments on the surface resistance R_s.

In Fig. 1(a), $R_s(T)$ is shown for two films of different thicknesses, deposited on different substrates. Data were taken at approximately 1-K intervals and the curves displayed in the figure represent the overall temperature dependence observed. In both cases R_s sharply decreases at T_c with no measurable temperature variation below about 75 K. Figure 1(b) shows the low temperature (below 70 K) variation of $R_s(T)$ for the two films of Fig. 1(a). Within our experimental resolution, R_s is unmeasurable below 70 K, and more detailed experiments employing superconducting cavities will be required to establish the upper limit of R_s at low temperatures. More precisely, the experiments indicate a variable but small residual loss [seen in Fig. 1(b)] at temperatures below approximately 70 K. The origin of this, and whether it can be separately interpreted as surface resistance has not been resolved. This residual loss has been subtracted from our result and we are focusing on the temperature-dependent part of R_s, $R_s(T)$.

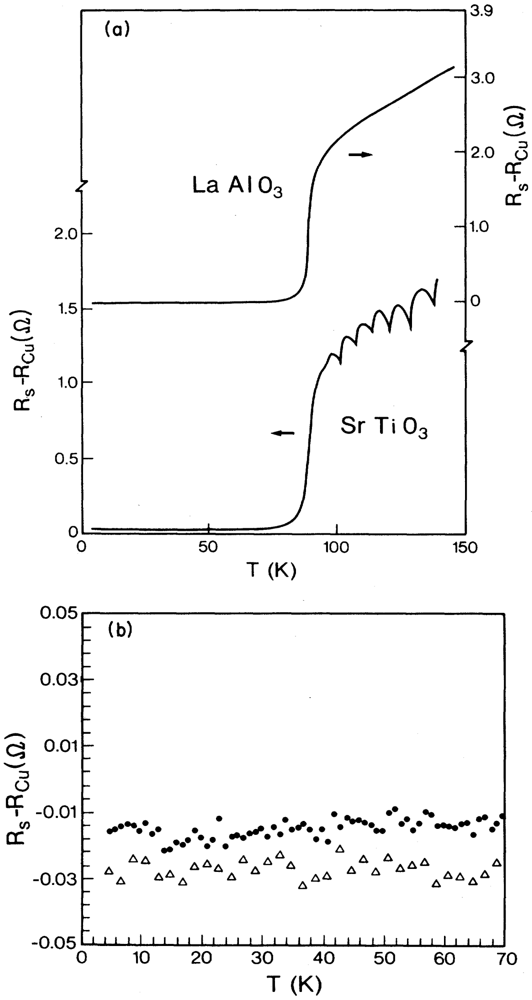

FIG. 1. (a) Temperature dependence of R_s-R_{Cu} in $YBa_2Cu_3O_7$ thin films on $LaAlO_3$ (4000-Å-thick) and $SrTiO_3$ (6000-Å-thick) substrates at 101.3 GHz. (b) Temperature dependence of R_s-R_{Cu} below 70 K for the films in (a). The triangles represent the 4000-Å-thick $LaAlO_3$ thin film and the circles represent the 6000-Å-thick $SrTiO_3$ thin film.

The film thickness is a factor of 2 to 3 smaller than the skin depth δ in the normal state

$$\left[\rho_n(100\,\mathrm{K})=50\,\mu\Omega\,\mathrm{cm},\quad \delta=\left(\frac{2\rho}{\mu_0\omega}\right)^{1/2}=1.1\,\mu\mathrm{m}\right]$$

and our experiments indicate substantial losses associated with leakage through the films in the normal state. First, the film with a smaller thickness, $d=4000$ Å, exhibits an apparent normal-state R_s value approximately twice that of the normal-state R_s measured on a thicker, $d=6000$ Å film. Both exceed the value which can be calculated using $R_s=(\mu_0\omega\rho_n/2)^{1/2}=0.45$ Ω for $\omega/2\pi=101.3$ GHz and $\rho_n=50$ $\mu\Omega$ cm. Using this value, and the measured R_s values at T_c, the amount of leakage can be estimated. Second, for films deposited on $SrTiO_3$ substrate, oscillations in $R_s(T)$ are seen. This has been shown[6] to be due to standing waves associated with the $SrTiO_3$ substrate which has a strongly temperature-dependent dielectric constant. This phenomenon can also be used to establish the amount of leakage.[6] Both observations indicate that while leakage is substantial in the normal state, in the superconducting state the relevant length scale is the penetration depth λ, and below about $0.98T_c$ negligible leakage is expected within our experimental error.

In Figs. 2–4, the superconducting surface resistance R_s, normalized to the normal-state surface resistance value

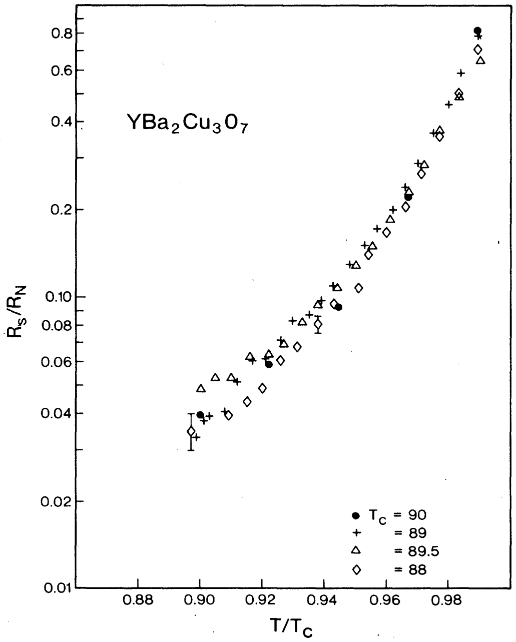

FIG. 2. Temperature dependence of the reduced surface resistance R_s/R_N vs reduced temperature T/T_c for the various $YBa_2Cu_3O_7$ thin films at 101.3 GHz. $R_N=0.45$ Ω corresponding to $\rho_n=50$ $\mu\Omega$ cm.

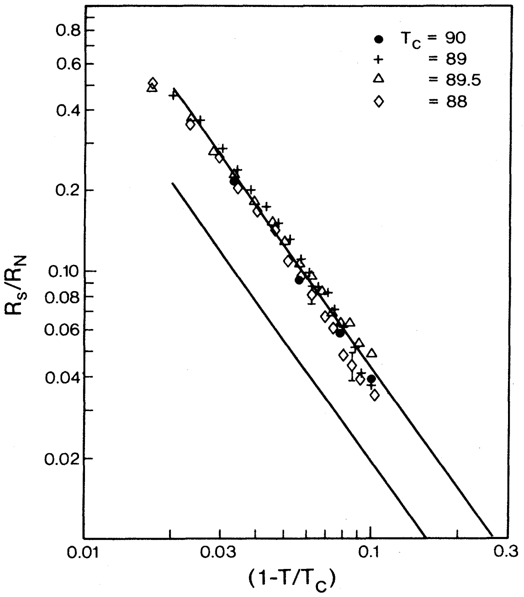

FIG. 3. Temperature dependence of the normalized surface resistance R_s/R_N vs reduced temperature $1-T/T_c$ of the various $YBa_2Cu_3O_7$ films at 101.3 GHz. The solid curves represent the predictions of the two-fluid model. The parameters used are $\sigma_n^{-1}=50$ $\mu\Omega$ cm and $\lambda(0)=1500$ Å for the lower curve and $\lambda(0)=1970$ Å for the upper curve. See text for detailed discussion.

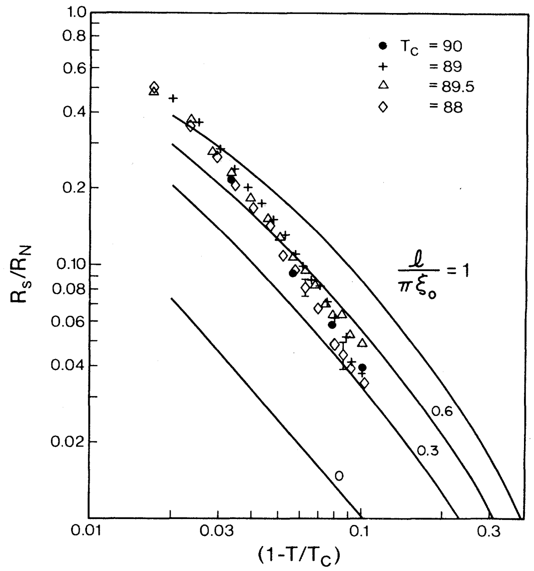

FIG. 4. Temperature dependence of the normalized surface resistance R_s/R_N vs reduced temperature $1-T/T_c$ of the various $YBa_2Cu_3O_7$ films at 101.3 GHz. The solid curves represent the predictions of the BCS theory with the ratio $l/\pi\xi_0=0$, 0.3, 0.6, 1.0, and $2\Delta(0)/k_BT_c=3.52$.

$R_N=0.45$ Ω at $T=T_c$, is displayed versus the reduced temperature T/T_c and $1-T/T_c$. Also included in Figs. 3 and 4 are several theoretical curves that will be discussed later. Since it is difficult to determine T_c accurately from our experiments due to leakage through the films near T_c, and no other independent method was used to determine T_c, we have treated it as a free parameter, and the values used are displayed in the figures. They are in agreement with the general trend of observing slightly reduced T_c values in films, as compared with single crystals. Several features of Figs. 2, 3, and 4 are of importance. First, the temperature dependence observed is similar for the three films measured, indicating an intrinsic behavior. Second, a specimen measured twice with a time difference of about one month with the sample stored in air at room temperature (points ● and +) shows little degradation, as evidenced only by a slight decrease of the transition temperature. We also note that R_s of the films investigated is significantly smaller than all other reported values (except for Ref. 8) that we are aware of.

The superconducting copper oxides differ from conventional superconductors in many ways. In addition to having significantly higher transition temperatures, they are anisotropic with the one-electron transfer in the *c* direction being much weaker than that in the *a* and *b* directions.[12] Furthermore, the coherence length in these materials is of the order of tens of angstroms[13] which is much shorter than the penetration depth of the magnetic field which is of the order of several thousand angstroms. This is in contrast with conventional clean superconductors in which the coherence length is typically much larger than the penetration depth.[14] For this reason, the electromagnetic response is described by a local London relation. Here we discuss two models for the surface impedance of a *c*-axis oriented film. First we consider a two-fluid model with

$$\sigma_1=\sigma_n\left(\frac{T}{T_c}\right)^4 , \tag{3}$$

and

$$\sigma_2=\frac{1}{\mu_0\omega\lambda^2(T)}=\frac{1-(T/T_c)^4}{\mu_0\omega\lambda^2(0)} . \tag{4}$$

For a *c*-axis normal oriented film, σ_n is the *ab* plane normal-state conductivity and $\lambda(0)$ is the zero-temperature *ab* penetration depth. Taking $\sigma_n=2\times10^6/\Omega\,\mathrm{m}$ corresponding to $\rho_n=50$ $\mu\Omega$ cm and $\lambda(0)=1500$ Å,[2,15] one finds that at 100 GHz, $\sigma_1/\sigma_2\ll1$ over the temperature region of interest,[16] so that Eq. (1) can be expanded, giving

$$\frac{R_s}{R_N}=\frac{1}{\sqrt{2}}[\mu_0\omega\sigma_n\lambda^2(0)]^{3/2}\frac{(T/T_c)^4}{[1-(T/T_c)^4]^{3/2}} . \tag{5}$$

In the temperature region near T_c where R_s was measured, this reduces to

$$\frac{R_s}{R_N}\cong6.06\times10^{-4}\left(1-\frac{T}{T_c}\right)^{-3/2} , \tag{6}$$

which is plotted as the lower curve in Fig. 3. If we take $\lambda(0)=1970$ Å we obtain the upper curve, which passes through the data.

The second approach that we have used to analyze these results is to calculate σ within the framework of the BCS theory. For a c-axis oriented film, we neglect the electron transfer between the planes, so that only the component of the wave vector of the electromagnetic field parallel to the layers enters. It is effectively zero for the micro- and millimeter waves of interest, so that the electromagnetic kernel relating the current density $\mathbf{j}$ to the vector potential A is evaluated with $\mathbf{q}=0$.[17] Thus the electromagnetic response is described by Eq. (1) with σ depending on $l/\pi\xi_0$, $2\Delta(0)/k_BT_c$, $\omega/\Delta(0)$, and T/T_c. Here l is the mean free path, which we assume to be limited by elastic scattering due to impurities, and ξ_0 is the zero-temperature coherence length $\hbar v_F/\pi\Delta(0)$. It is of course an open question as to whether such a BCS analysis is appropriate and in particular whether the scattering lifetime, which probably arises from the underlying dynamics, can be approximated by an elastic scattering mean free path even over the relatively narrow temperature range of this experiment.

Results obtained from the BCS theory with $2\Delta(0)/k_BT_c=3.52$ are compared with the experimental data in Fig. 4. In the dirty limit $l/\pi\xi_0\ll 1$, the conductivity which enters Eq. (1) is given by the Mattis-Bardeen result[18] appropriate for our local electrodynamics. The temperature dependence of R_s/R_N in the dirty limit is shown as the lowest curve in Fig. 4. An estimate of l/ξ_0 can be obtained from

$$\sigma_n\lambda_L^2(0)=\frac{\tau}{\mu_0}=\frac{l}{\pi\xi_0}\left(\frac{k_BT_c}{2\Delta(0)}\right)\frac{2\hbar}{\mu_0k_BT_c}\,. \qquad (7)$$

Taking $\sigma_n^{-1}=50\ \mu\Omega\,\text{cm}$, $T_c=90$ K, $2\Delta(0)/k_BT_c=3.52$, and $\lambda_L\cong 1500$ Å gives[19] $l/\pi\xi_0\cong 1.2$. Results for various values of $l/\pi\xi_0$, keeping $2\Delta(0)/k_BT_c=3.52$, are shown as the other solid curves. Even for the best case, $l/\pi\xi_0=0.6$, we see that the fit is not as good as that of the two-fluid model. The calculated BCS curve has a smaller slope than the experimental data. We note that for larger values of $2\Delta(0)/k_BT_c$, the BCS curve decreases more rapidly as T/T_c decreases, and the fit is slightly better. At present we believe that better data on both R_s and X_s, particularly at low temperatures, is required for a meaningful comparison with a stronger-coupling theory in which $2\Delta(0)/k_BT_c$ is larger.

In summary, we have measured the temperature dependence of the surface resistance of several $YBa_2Cu_3O_7$ superconducting thin films on different substrates. Our results are compared with those of the two-fluid model and the BCS theory, including the mean-free-path effect. The two-fluid model can account for the slope, but to have the correct magnitude with $\rho_n=50\ \mu\Omega\,\text{cm}$ at T_c, it requires $\lambda(0)\cong 1970$ Å, which is somewhat larger than the currently accepted value of the zero-temperature penetration depth $\lambda(0)=1500$ Å. On the other hand, the BCS theory can account for the correct magnitude of R_s/R_N using $2\Delta(0)/k_BT_c=3.52$ and a reasonable value of $l/\pi\xi_0=0.6$. However, it predicts a slower decrease in R_s/R_N as the temperature is reduced than that which is observed. It will be interesting to make a similar study of the microwave reactance, which is currently in progress.

One of us (J.J.C.) would like to thank Superconducting Technologies Inc. for their hospitality when part of the work was performed at their facilities. We would like to thank F. Gamble and R. Hammond of Superconducting Technologies Inc. for useful discussions. Work at University of California, Los Angeles (UCLA) was supported by the UCLA Consortium on High Temperature Superconductivity.

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