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Microwave transmission through films of $YBa_2Cu_3O_{7-\delta}$

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We report measurements of the magnitude and phase of microwave transmission through thin films (1 μm) of the 90-K superconductor $YBa_2Cu_3O_{7-\delta}$ on Al_2O_3, MgO, and ZrO_2 substrates. Real and imaginary parts of the conductivity, σ_1 and σ_2, were evaluated from the measurements by comparing with a calculation of the transmission for our experimental configuration. Both parts of the conductivity show qualitative agreement with Bardeen-Cooper-Schrieffer type conductivities. From σ_1, we find a range of gap values $2\Delta/k_BT_c=1.3$ to 6.5. From the measured σ_2 the magnetic penetration depth can be evaluated.

Microwave measurements on superconductors can be utilized to evaluate magnetic penetration depths as well as to obtain information on the mechanism of superconductivity from the temperature dependence of the complex conductivity or surface impedance. There have been several reports of such measurements on $YBa_2Cu_3O_{7-\delta}$. Recent examples are the publications by Carini, Awasthi, Alavi, and Grüner on ceramics[1] and on oriented films,[2] and those of Porch, Waldram, and Cohen[3] on powders. All reported measurements on high-temperature superconductors have utilized cavity perturbation techniques which, except in the case of powders, measure surface impedances. The drawbacks of such methods are that the sample must be entirely in the electric or the magnetic field; otherwise the analysis becomes complex. In addition, because the surface impedance is dependent upon the penetration depth, it becomes a complicated process to extract the conductivity. For experimental geometries which utilize a thin-film sample as one cavity wall, losses due to leakage through the film can dominate the resistive loss in the film.[2] We have chosen instead to measure the magnitude and phase of microwave transmission *through* superconducting films.[4,5] From these measurements the complex conductivities are evaluated *directly*.

Our measurements were made on thin (of order 1 μm) films of $YBa_2Cu_3O_{7-\delta}$ on crystalline Al_2O_3, MgO, and Y stabilized ZrO_2 substrates. The substrates were generally between 0.02 and 0.11 cm thick. The films were deposited in a system consisting of three electron beam heated sources containing Y, Ba, and Cu in an oxygen partial pressure of 0.1 to 1 mTorr. Substrate temperatures during the deposition were generally about 375 °C but films could be made at temperatures from room temperature to about 700 °C. Post-deposition annealing of the films at a temperature of about 925 °C in flowing oxygen was required to form the high-T_c material. Additional details of the fabrication process can be found in Ref. 6.

These samples were generally polycrystalline with grain sizes up to several microns. Low-frequency (10 Hz) resistive transitions for these films were generally between 80 and 85 K. Epitaxial films that show high critical current and T_c close to 90 K could be produced on single crystal, <100>-oriented $SrTiO_3$ substrates.[7] However, good microwave transmission measurements were not possible due to the large magnitude and temperature dependence of the dielectric constant of $SrTiO_3$.

Our experimental technique was essentially the same as that of Ref. 4. The film-substate combination was held in a frame which was suspended across the x-band waveguide between two waveguide flanges. The U-shaped waveguide was placed in a helium cryostat with both input and output ends at the top. Microwave power at 9 GHz was split between the input and a reference signal. The measurements were made by reading the attenuation and phase required to null the reference against the transmitted output. With a copper foil replacing the film, the leakage level was less than -70 dB; however, leakage through microcracks or pinholes in the films was greater than this value. (This is discussed below in more detail.) Background attenuation and phase, obtained from measurements of transmission as a function of temperature in the absence of the sample, were subtracted off. Measurements made on the substrates alone were used to determine any temperature dependence of the dielectric constants.

The conductivity,[8] $\sigma=\sigma_1+i\sigma_2$, was evaluated from the data by comparison with a theoretical expression for the transmission through a superconducting film which is homogeneous, isotropic, nonmagnetic, and on a dielectric substrate. In keeping with the observation that the transmission through some of our films was less than -30 dB, we have not made thin-film assumptions in deriving our expression. This is a departure from Refs. 4 and 5 where the film thickness was assumed to be small compared with the magnetic penetration depth λ and in fact, the smallest λ we measured was only twice the film thickness. The experiment uses a TE_{01} mode propagating in the waveguide. In an exact calculation for the TE_{01} mode, the longitudinal component of the magnetic field must be included in matching the boundary conditions. However, in our calculation we assume a plane wave normally incident on the film and only account for the mode by utilizing TE_{01} wave vectors. The transmission amplitude τ and phase θ are defined through the ratio of the transmitted field to the incident field which is given by the following expression, assuming an $e^{-i\omega t}$ time dependence:

$$\tau e^{i\theta}=\frac{E_4}{E_1}=8e^{-ik_1(d+l)}\{e^{-ik_2d}(1+Z_{12})[e^{-ik_3l}(1+Z_{23})(1+Z_{34})+e^{ik_3l}(1-Z_{23})(1-Z_{34})]+e^{ik_2d}(1-Z_{12})[e^{-ik_3l}(1-Z_{23})(1+Z_{34})+e^{ik_3l}(1+Z_{23})(1-Z_{34})]\}^{-1} \quad (1)$$

d=superconducting film thickness, l=substrate thickness, k_j is the complex wave vector in region j, and Z_{ij} is an impedance ratio equal to k_j/k_i. The subscripts reference the propagation regions as follows: 1=incident waveguide; 2=superconductor; 3=substrate; and 4=exit waveguide ($k_4=k_1$). For the TE_{01} mode $(k_j)^2=(k_{0j})^2-(\pi/a)^2$, where k_0 for each region is the corresponding infinite plane wave value, and a is the width of the guide. The dispersion relation inside the superconductor was solved for by adding a supercurrent term to the generalized Ampère's law of Maxwell's equations in addition to the term for a normal current. Taking the curl of both sides of this equation allows insertion of London's postulate and relates λ, and, hence, σ_2, to τ and θ in a natural way. The imaginary part of the conductivity in the London theory is defined as

$$\sigma_2\equiv(\mu\omega\lambda^2)^{-1}, \quad (2)$$

where μ is the permeability of free space.

At temperatures T, above T_c, it is a simple matter (see Ref. 4) to evaluate the normal surface resistance, R_n. (The microwave values were within a factor 2 of the R_n from dc measurements.) However, in order to evaluate the conductivities from the measured transmission amplitude and phase below T_c, the transcendental equations which result from inverting Eq. (1) must be solved numerically. A considerable simplification results if Eq. (1) is cast in terms of σ_1/σ_c and σ_2/σ_c (here and in the following, the subscript c refers to values at $T=T_c$), the measured R_c is used, and the transmission is normalized to that at T_c; i.e., after solving for R_n above T_c we then solve for the relative conductivities below T_c from the measured $\tau(T)/\tau_c$ and $\theta(T)-\theta_c$. For this purpose T_c was arbitrarily taken to be the temperature at the maximum of $d\tau/dT$.

Figure 1(a) shows the temperature dependence of the measured power transmission normalized to T_c for three separate runs on a film deposited on Al_2O_3. The relative phase, $\theta-\theta_c$, is also shown. (For clarity on the figure we have plotted $\theta_c-\theta$, i.e., the negative of the phase change.) Each run was started at room temperature and the data taken during cooling. While the different sets of data are in good agreement at higher temperatures, the departures at low temperature are significant. In particular, note that on run No. 1 the transmission goes through a minimum at about 30 K; on the other two runs both transmission and phase tend to level out below about 50 K. These behaviors and their variations with temperature cycling can be explained by assuming that there is a small amount of leakage transmission through the films which, as the temperature is lowered, eventually dominates the decreasing transmission through the superconducting material. Possible sources of leakage are microcracks,[9] pinholes or normal regions. The number and distribution of cracks would be affected by stresses caused by expansion and contraction of the sample when its temperature is cycled and, therefore, this seems the most probable source of leakage.

In Fig. 1(b), we show the normalized conductivities $\sigma_{1,2}/\sigma_c$ obtained from the data of Fig. 1(a) using Eq. (1). The effect of leakage is reflected here as a leveling off of σ_2 at low temperatures. Also, film deterioration with cycling and/or exposure to air and water vapor is evidenced by the smaller values of σ_1 and σ_2 for runs 2 and 3 than for run 1.

The principal qualitative features to be noted in Fig. 1(b) are the peak in σ_1 below T_c, and the monotonic increase in σ_2 with decreasing temperature below T_c. These types of behavior are characteristic of a superconductor which for decreasing temperatures exhibits both an increasing energy gap and increasing density of quasiparticle states at the gap edge as, for example, a BCS superconductor. The peak is not related to the "coherence peak" observed in attenuation of high-frequency ultrasonics. It is simply due to competition between the increasing density of states and the decreasing number of quasiparticles. Later, we explain how the σ_1 peak can be used to evaluate a zero-temperature BCS gap parameter $2\Delta_0$ from Mattis-Bardeen theory;[10] in the present case we find $2\Delta_0=1.26k_BT_C$.

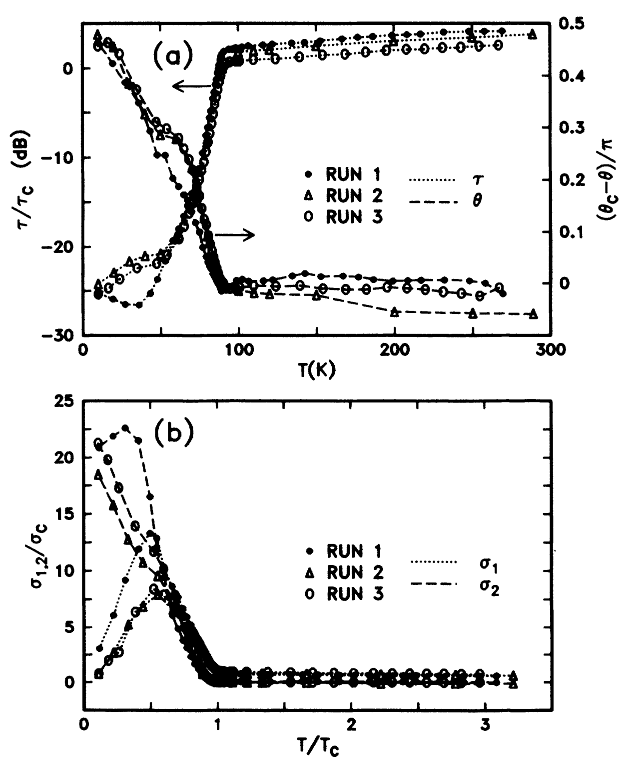

FIG. 1. (a) Relative amplitude τ/τ_c and the negative of the phase change $\theta_c-\theta$ for a film on Al_2O_3. Three runs explained in the text are shown. (b) Real and imaginary parts of the conductivity obtained from the data in (a).

From Eq. (2), using the measured $R_c = 42\,\Omega/\square$, $d = 1.1\ \mu$m, and $\sigma_2/\sigma_c \simeq 20$, we find $\lambda \simeq 2.5\ \mu$m at 30 K. Because of the polycrystalline and inhomogeneous nature of the film, this value of λ should be viewed as an upper limit. In a polycrystalline sample, where the constraints of orientation are absent, the low conductivity regions will tend to dominate in an averaged measurement of the penetration depth. Magnetization measurements[11] on single crystals give highly anisotropic values for the zero-temperature penetration depth, with the largest value reported equal to 0.8 μm.

The transitions were sharper for films on MgO and ZrO_2 substrates. However the leakage was larger, limiting the results to higher temperatures than for the films on Al_2O_3. A possible explanation for this difference[12] is that diffusion of Al into the film, which is known to occur during the high-temperature annealing step, produces a more uniform and mechanically stronger interface.

Figure 2(a) shows the data from a 1.2 μm film on ZrO_2. As in Fig. 1(a) the transmission goes through a minimum but here the phase change becomes larger than $\pi/2$. Since $\pi/2$ is the largest phase allowed by Eq. (1), it is clear that the leakage signal dominates transmission through the film and, as indicated in Fig. 2(b), solutions of Eq. (1) for the conductivities could not be found for temperatures below 70 K. Nevertheless, even within this limited temperature range, the peak in σ_1 and rapid increase in σ_2 below T_c are clearly evident. From the σ_1, we find $2\Delta_0 = 3.6 k_B T_c$, which within experimental error equals the BCS value $3.52 k_B T_c$. However, as above, there is not quantitative agreement with the BCS temperature dependence as shown by the theoretical σ_1, evaluated from Ref. 10 and plotted in Fig. 2(b). For the parameters of this film, $R_c = 12.5\,\Omega/\square$, $d = 1.2\ \mu$m, and maximum $\sigma_2/\sigma_c \simeq 7$, we find $\lambda \simeq 2.6\ \mu$m at 70 K. The equality of this value with the 30 K value from Fig. 1 is coincidental.

A total of 21 runs were made on six films, two on each of the three substrates. One of the films on ZrO_2 had an extremely broad transition and showed only a slow monotonic increase in both σ_1 and σ_2 with no σ_1 peak. The other five samples all exhibited qualitative BCS-type behavior for the temperature dependence of the conductivities, but quantitative agreement with BCS was not obtained. However, *if one assumes BCS theory to apply,* the conductivities given by Mattis and Bardeen can be evaluated with $2\Delta_0$ as a parameter. Then the value of T/T_c at which the maxumum in σ_1 occurs, $(T/T_c)_p$, is a single-valued function of $2\Delta_0/k_B T_c$. (This maximum only occurs for $\hbar\omega \ll 2\Delta_0$.) This functional dependence can then be used to evaluate $2\Delta_0/k_B T_c$ from the measured (T/T_c). Gap values thus obtained from 14 runs (five samples) are plotted in Fig. 3. They are distributed over the range $2\Delta_0/k_B T_c = 1.3$ to 6.5. This is in rough agreement with the spread of values observed in ceramic samples by infrared reflectivity,[13] NMR,[14] and tunneling measurements.[15–18] However, our values can only be considered as lower limits because the breadth of the transitions due to the inhomogeneous, polycrystalline nature of the films tends to move the σ_1 peaks to lower temperatures thereby also decreasing the measured gaps.

In conclusion, we have shown that the microwave conductivity of $YBa_2Cu_3O_{7-\delta}$ exhibits the qualitative behavior expected for a BCS-type superconductor. We have also shown that a BCS gap can be calculated from measurements of σ_1. The magnetic penetration depth was determined from σ_2. However, the accuracy of our measurements of conductivity was limited by the presence of microwave leakage through the films, and the latter also indicates that it may be possible to utilize microwave measurements as a sensitive test of film quality and mechanical properties.

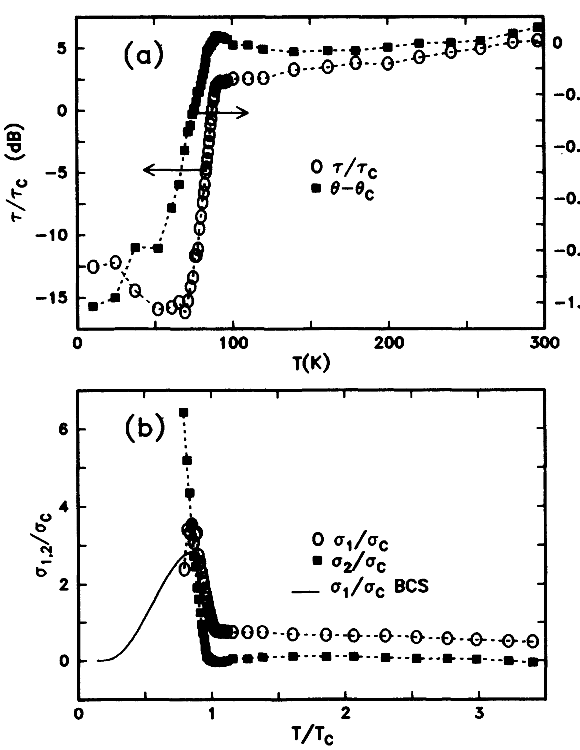

FIG. 2. (a). Relative transmission and phase change measured on a film deposited on ZrO_2. (b) Conductivities obtained from the data in (a). Solid line is the real part of the conductivity calculated from Ref. 10.

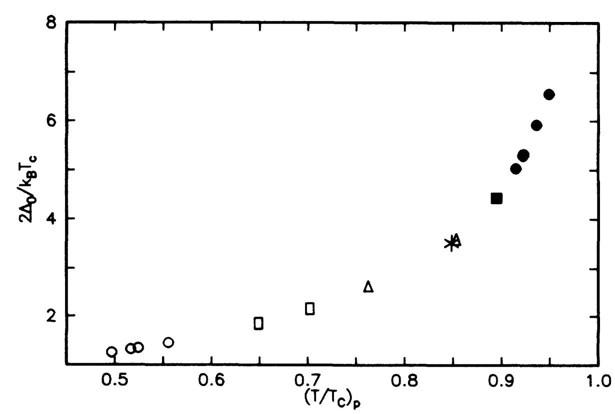

FIG. 3. Gap values evaluated from measured temperatures at the σ_1 peak, as explained in the text, are shown by symbols for five samples. The asterisk (*) indicates the point at $2\Delta_0 = 3.52 k_B T_c$.

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