Millimeter-wave complex-conductivity measurements of Bi-Ca-Sr-Cu-O superconducting thin films

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The first experimental determination of the complex conductivity $\sigma_1(T)$ and $\sigma_2(T)$ of superconducting oriented polycrystalline thin films of Bi-Ca-Sr-Cu-O has been made at 60 GHz. A sharp rise in σ_1 is observed below T_c. The microwave data can be fit by a model of superconducting grains nucleating in a normal conducting matrix. The penetration depth at 40 K deduced from this model is 1.3 μm.

Determination of the millimeter-wave complex conductivity of the recently discovered high-T_c superconductors can yield fundamental material properties that are critical both for high-frequency device applications and for basic understanding of the superconducting mechanisms in these new classes of materials. To date, reported data have been restricted to millimeter-wave surface resistance determinations on thin films[1] and cavity perturbation results on bulk ceramics.[2] These measurements are insufficient to unambiguously separate the magnitude of the real and imaginary components of the complex conductivity $\sigma^* = \sigma_1(\omega,T) - i\sigma_2(\omega,T)$. Measurements of the amplitude and phase of a transmitted signal through a thin film provide the most direct determination of σ_1 and σ_2. Alternatively, simultaneous measurements of reflected and transmitted power[3] or measurements of transmission supplemented by a suitable model[4] can be used.

All the film measurement methods are sensitive to the substrate on which the film is deposited. For high-T_c film growth, the most preferred substrate material is $SrTiO_3$, which has a ferroelectric phase transition in the same cryogenic temperature range as the new superconductors. The large temperature dependence in its dielectric properties makes it exceedingly difficult to isolate the transmission properties of the deposited thin film.

Recently, we have been successful in the growth of highly oriented polycrystalline bismuth-calcium-strontium-copper-oxide (BCSCO) thin films on single-crystal MgO substrates. MgO is ideally suited for transmission-type measurements since its millimeter-wave loss tangent is less than 10^{-4}, while its dielectric constant [9.73 at room temperature (RT)] changes by only approximately 2% over the cryogenic temperature range of interest. Consequently, the contributions to the transmission properties from the substrate layer can easily be accounted for.

This paper describes the results of a study undertaken on several BCSCO thin films near 60 GHz. The measurement method uses quasioptical techniques to produce a near-diffraction-limited beam that can be focused onto a sample mounted in a holder on a cryogenic cold stage. The experimental apparatus is shown in Fig. 1. The system consists of a set of spot focusing lens antennas with a diameter of 15.25 cm and an f number of unity, suitably positioned such that the sample is at the focal plane of the antennas. The spot size (full width, half power) of the focused linearly polarized beam is of the order of 0.9 cm at 60 GHz. The vacuum windows were made of 0.070 in. Plexiglas to minimize standing waves and stray reflections.

The power transmission coefficient at a given frequency is measured by first obtaining a background transmission with the sample removed from the holder and with the chamber evacuated. Power levels are calibrated with precision rotary-vane attenuator no. 1. The sample is then placed in the holder and the with-and-without sample difference recorded as a function of temperature. The sample and window orientations are adjusted with respect to the incident microwave beam to give lowest standing waves and minimum stray radiation at the detector. In addition, microwave absorbers are used both within and around the vacuum chamber to further reduce multiple reflections. The transmitted power is measured with a crystal detector and a lock-in amplifier operating at an amplitude-modulation frequency of 1 kHz superimposed on the incident signal by a ferrite modulator. The dynamic range of the system is approximately -65 dB. The residual stray radiation, as determined by replacement of the sample with an identically sized metal plate, is approximately -60 dB.

The change in phase of the transmitted signal through the sample is determined by balancing the microwave signals in the sample and reference arms of the phase bridge with precision rotary-vane attenuator no. 2, and tuning for a null with the reference phase shifter. The attenuator setting is essentially phase independent, the relative change in phase being on the order of 1° over attenuation range 0–40 dB. The system has a phase resolution of $\pm 1°$ over the sample insertion loss range of 0 to -50 dB.

The BCSCO superconducting films were fabricated by reactive ion-beam deposition at room temperature on single-crystal 1.3×1.3 cm (001) MgO substrates and then annealed to 880 °C in oxygen. X-ray diffraction showed the films to be in the superconducting orthorhombic structure and oriented with the c axis perpendicular to the substrate surface. Typical critical currents are in the range 5×10^4 A/cm^2 at 10 K. Details of the fabrication procedure and other physical and chemical properties of these films are given in a separate publication.[5]

For the millimeter-wave measurements, 0.5- and 0.1-

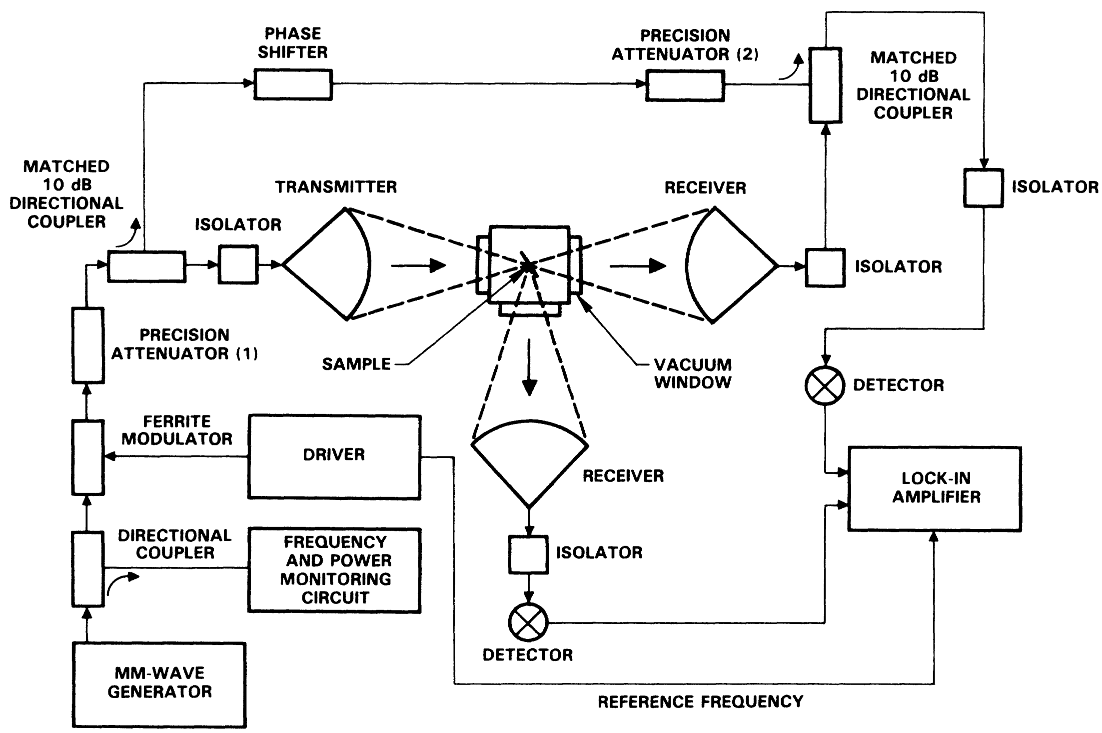

FIG. 1. Schematic of measurement apparatus.

μm films with similar dc properties were studied primarily at 62.4 GHz. The temperature dependence of the dc resistivity ρ_{dc} is shown in Fig. 2(a) for the 0.1-μm film. The critical temperature (T_c), as determined by the vanishing of ρ_{dc}, is 68 K. For comparison, the millimeter-wave transmission obtained at 62.4 GHz for the two films is also shown. In the region well above T_c, the transmission loss follows the observed trend of the dc resistivity. The transmission drops abruptly near T_c, similar to the behavior seen in films of conventional type-I or type-II superconductors.[3] However, the width of the observed transition is much broader. The corresponding measured relative phase shift for these two cases is shown in Fig. 2(b). There is essentially no phase shift over the temperature range 100–300 K. At lower temperatures, a large negative phase shift gradually develops, but the total never reaches the $-\pi/2$ value expected for a fully superconducting thin film with $\sigma_2 \gg \sigma_1$.

Analysis of the transmission and phase data of the type shown in Fig. 2, with known dielectric properties assigned to the substrate material, yields directly the complex conductivity of the film. Results of this analysis are shown in Fig. 3. In Fig. 3(a), the real dielectric constant $\epsilon' = \sigma_2/\varepsilon_0\omega$ (where ε_0 is the free-space permittivity and ω the angular frequency) for the 0.1-μm film at two different frequencies (58.2 and 62.4 GHz) is compared with that for the 0.5-μm film at 62.4 GHz. The values in all cases are close to zero above T_c, which is characteristic of an ordinary conductor. Below T_c, the dielectric constant shows a steady decrease with no indication of a low-temperature saturation. Similar comparisons are made in Fig. 3(b) for the computed ac conductivity (σ_1). The magnitudes and temperature dependences above 90 K are in excellent agreement with the observed dc conductivity. At temperatures below 90 K, however, the data show a conductivity increase reaching a broad peak which is approximately three times larger than the normal-state conductivity σ_n at 90 K, followed by the beginning of a decreasing trend at lower temperatures. The two frequencies of measurement are sufficiently close that we were not

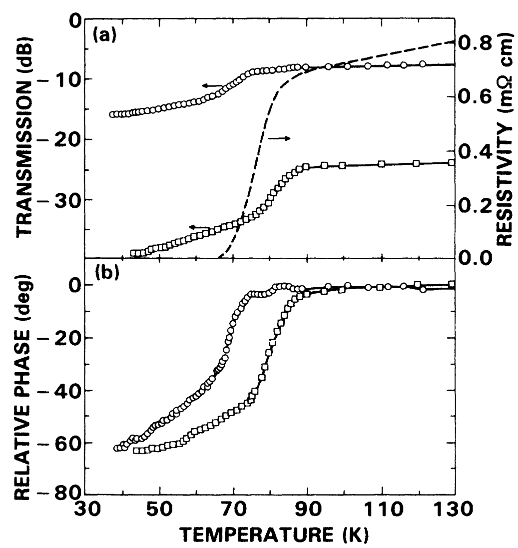

FIG. 2. (a) Measured dc resistivity for a 0.1-μm-thick BCSCO film on single-crystal MgO substrate. Also shown are the observed transmission loss at 62.4 GHz for 0.5- (square) and 0.1-μm (circle) BCSCO films. (b) Measured corresponding phase shifts for the same films.

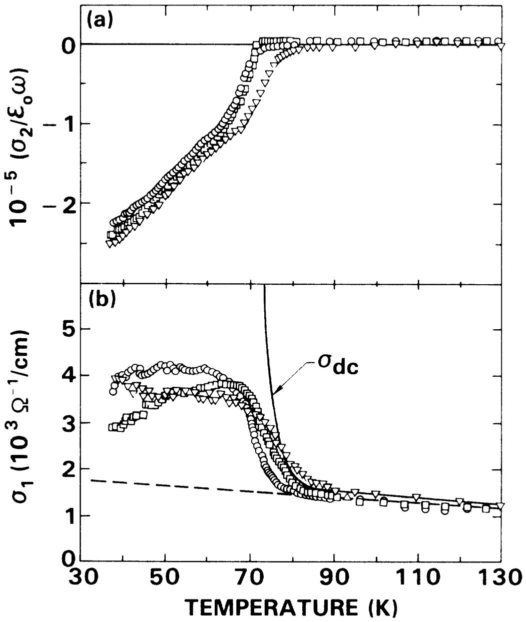

FIG. 3. (a) Computed values for $(\sigma_2/\varepsilon_0\omega)$ of the 0.5-μm BCSCO film at 62.4 GHz (triangle) and the 0.1-μm BCSCO film at 62.4 GHz (circle) and 58.2 GHz (square). (b) Computed corresponding σ_1 for the same films. Shown for comparison is the measured dc conductivity of the 0.1-μm BCSCO film.

able to reliably determine a frequency dependence for either σ_1 or σ_2.

The trends in microwave properties below T_c found in these films may reflect either temperature-dependent intrinsic properties of a homogeneous superconductor or mixture effects arising from superconducting regions growing in a normal conducting matrix. The most striking feature of our data is the large increase in σ_1 below 90 K. BCS theory for a homogeneous superconductor with an isotropic energy gap predicts such a behavior,[6] but it is necessarily accompanied by an even larger increase in nuclear spin relaxation rate $1/T_1$. In fact, measurements of this rate on the closely related high-T_c oxide $YBa_2Cu_3O_{7-\delta}$ show that $1/T_1$ decreases rapidly and monotonically below T_c.[7] This apparent discrepancy can be removed if we adopt a mixture model in which each superconducting grain is assumed to possess large negative ϵ' and a rapidly decreasing σ_1/σ_n, following the observed trend in spin relaxation rate. The exclusion of the microwave electric field from these localized regions of large negative dielectric constant will raise the average field and current in the surrounding normal matrix, causing an apparent conductivity increase in the composite film.

We have used this picture to deduce the intrinsic superconducting dielectric properties of the isolated grains and their volume fraction ϕ_s from the measured film data of Fig. 3. The oriented, platelike microstructure of BCSCO films suggests that planar superconducting inclusions, nucleating and growing in a homogeneous planar sheet, should be an appropriate model.[5] Specifically, we have used a mixture rule for dielectric properties in the plane of the film which has the two-dimensional Bruggeman[8] form at high volume fractions ($\phi_s \geq 0.5$), i.e.,

$$\phi_s = 1 - [(\epsilon_n/\epsilon)^{1/2}(\epsilon_s - \epsilon)/(\epsilon_s - \epsilon_n)] \, ,$$

and the corresponding Maxwell Garnett[8] form at low volume fractions ($\phi_s < 0.5$), i.e.,

$$\phi_s = (\epsilon - \epsilon_n)(\epsilon_s + \epsilon_n)/(\epsilon_s - \epsilon_n)(\epsilon + \epsilon_n) \, .$$

Here s and n on the complex dielectric constant ϵ (where $\epsilon = i\sigma^*/\varepsilon_0\omega$) denote the superconducting grains and the normal matrix, respectively, while the composite film dielectric constant is unlabeled. Of the seven variables appearing in these expressions (ϕ_s; σ_1^s, σ_2^s; σ_1^n, σ_2^n; σ_1, σ_2), only the last pair is directly determined from the measurements. In order to compute ϕ_s and σ_2^s, we have extrapolated the linear behavior of σ_1^n and σ_2^n to the temperature range below 90 K (see Fig. 3) and made the assumption that σ_1^s is negligible compared to σ_1^n. Any weak-link effects[9] within grain clusters are lumped into the equivalent ϵ_s for the clusters.

Figure 4(a) shows the volume fraction ϕ_s deduced from the above model as a function of temperature for the 0.1-μm film at 62.4 GHz. Results for the other sets of data exhibit the same reasonable trend in the growth of ϕ_s with decreasing temperature. The dc resistance of the film is found to vanish near $\phi_s = 0.5$, indicating the formation of continuous superconducting paths at this volume fraction.

The corresponding trend in σ_2^s of the superconducting grains is shown as $\epsilon_s' = (\sigma_2^s/\omega\varepsilon_0)$ in Fig. 4(b). The assumption of negligible σ_1^s should be best at the lower temperatures, and there one finds ϵ_s' to be approximately constant. The obtained value of -3.6×10^5 corresponds to a London penetration depth of 1.3 μm, which is an order of magnitude larger than the static field penetration depth determined for the $YBa_2Cu_3O_{7-\delta}$ material.[10] At temper-

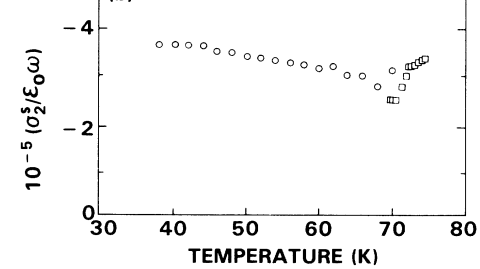

FIG. 4. (a) Computed superconducting volume fraction as a function of temperature for the 0.1-μm BCSCO film by using the Bruggeman (circle) and Maxwell Garnett (square) two-dimensional effective medium models. (b) Computed corresponding $\sigma_2^s/\varepsilon_0\omega$ of the superconducting grains.

atures near 75 K, the fitted values of ϵ_s' depart from their trend toward smaller magnitude, which may reflect the inadequacy of our assumed model. In this range, σ_1^s should be growing rapidly toward σ_n and may no longer be negligible. It should also be noted that the phase measurements have a large percentage uncertainty above 70 K, so that there is a correspondingly larger uncertainty in ϵ_s'.

The essential conclusions of this work may be summarized as follows: (1) A direct millimeter-wave determination of both σ_1 and σ_2 for thin BCSCO films on MgO has been achieved by the simultaneous measurement of transmitted amplitude and phase at cryogenic temperatures. (2) A factor of 3 rise in film conductivity below 90 K has been observed, accompanied by a steady decrease in film permittivity to large negative values. (3) While these features are possibly intrinsic properties of a homogeneous superconducting film, we propose that they are caused by isolated superconducting regions nucleating in a normal matrix as the temperature is decreased. (4) Analysis of the film data based on this model gives a conventional saturating behavior for ϵ_s' at low temperatures and a reasonable trend in the volume fraction occupied by the superconducting component.

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