Millimeter-wave surface resistance measurements in highly oriented $YBa_2Cu_3O_{7-\delta}$ thin films

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We have measured the temperature dependence of the millimeter-wave surface resistance $R_s(T)$ in two oriented polycrystalline thin-film samples of $YBa_2Cu_3O_{7-\delta}$. Below 80 K, R_s drops rapidly as the temperature is decreased as expected for a superconductor, but excess surface resistance is observed at low temperatures in contrast to the predictions of the Bardeen-Cooper-Schrieffer theory. The measured temperature, frequency, and orientation dependence of the surface resistance are presented and discussed.

Measurement of the surface resistance of the new oxide superconductors is of fundamental physical and technical interest. For understanding the physical origins of high-temperature superconductivity, these measurements provide information about the energy gap, its anisotropy, and the presence or absence of nodes in the energy gap $\Delta(k)$, such as are associated with some exotic pairing mechanisms. On the technical side, they provide an extremely sensitive test of the quality of the materials. In addition, the magnitude of the surface resistance has intrinsic technical importance, especially for applications in the millimeter-wave range of the spectrum and beyond.

Early measurements of $R_s(T)$ at 100 GHz in $La_{2-x}Sr_xCuO_{4-\delta}$ samples have suggested a conventional thermally activated temperature dependence for the surface resistance at low temperatures, but the results were heavily influenced by sample quality.[1] Measurements at 9.85 GHz in $YBa_2Cu_3O_{7-\delta}$ have shown both interesting similarities to, and departures from, the predictions of Bardeen-Cooper-Schrieffer (BCS) theory,[2] but again, were made on poorly characterized ceramic specimens.

In this work, we report measurements of the surface resistance of two oriented polycrystalline $YBa_2Cu_3O_{7-\delta}$ films, one predominantly with the a axis and one with the c axis perpendicular to the substrate. We will compare the temperature dependence of the surface resistance $R_s(T)$ at 102 GHz in the films to search for anisotropy. We have also measured the c axis film at 148 GHz to make an initial determination of the frequency dependence of the surface resistance.

The surface impedance Z_S of a superconductor with $\lambda \gg \xi_0, l$ (where λ is the penetration depth, ξ_0 is the coherence length, and l is the electron mean free path) measured at frequency ω is related to the complex conductivity $\sigma = \sigma_1 - j\sigma_2$ by

$$Z_S = [(j\omega\mu_0)/(\sigma_1 - j\sigma_2)]^{1/2} . \qquad (1)$$

The losses are proportional to the real part of Z_S, which is the surface resistance R_S. In the limit where $\sigma_1 \ll \sigma_2$ (typically, at low temperatures compared with T_c and with $h\omega \ll k_B T_c$), the surface resistance is approximately

$$R_S \approx \tfrac{1}{2} (\omega\mu_0/\sigma_2)^{1/2} \sigma_1/\sigma_2 , \qquad (2)$$

and $\sigma_2 \approx 1/(\omega\mu_0\lambda^2)$. Therefore, R_s is proportional to λ^3, and for a BCS-like superconductor, $\sigma_1 \propto \ln(\Delta/h\omega) \times \exp(-\Delta/k_B T)$,[3] so the predicted frequency dependence for R_S is proportional to $\omega^2 \ln(\Delta/h\omega)$. The ω^2 dependence of the loss in a superconductor reflects the inductive response of the superfluid and is essentially independent of the microscopic details of the superconductivity. In contrast, σ_1 depends intimately on such details. We note that although the ω^2 dependence for the surface resistance is characteristic of a superconductor, other nonintrinsic losses, such as those produced by a thin, normal-metal surface layer or by grain boundaries, may have a similar frequency dependence.

The thin films were prepared by reactive magnetron co-sputtering from three metal targets in an oxygen background onto {100} $SrTiO_3$ substrates. The films are disordered as deposited. Epitaxial films result after high-temperature oxygen annealing.[4] The compositions of the films were very close to the 1:2:3 stoichiometry, with a compositional variation across the film of about 1 at.%. The films were polycrystalline.

The samples used in the surface impedance measurements were selected on the basis of their x-ray diffraction patterns and critical current densities. Measurements of the resistive transitions were not made prior to the surface impedance measurements since electrical contacts would have damaged the film surfaces. Similar films have transition widths as narrow as 2 K with full transitions at 88 K. Critical current densities were deduced from magnetization hysteresis loops as described by Bean.[5]

The first film was essentially completely a-axis oriented, 2.5-μm thick, and had a critical current density, J_c, of 1.2×10^5 A/cm^2 at 4.2 K. The second film was 70% c-axis–30% a-axis oriented, 1-μm thick, and had $J_c = 4 \times 10^6$ A/cm^2 at 4.2 K. The measured critical current densities at 4.2 K differ by more than a factor of 30 between our two samples, which is about three times larger than the factor of 10 anisotropy that has been found in similar measurements on single crystals.[6] Surface studies, such as Rutherford backscattering (RBS),[7] surface analysis by laser ionization (SALI), and Auger measurements, all reveal that the surface of similar films are not the 1:2:3 phase. For samples on $SrTiO_3$ substrates similar to those measured here, the RBS results show that virtually no impurities have been introduced into the film via reaction with the substrate. The actual surface layer depth and composition appears to vary widely from sample to sample and depend on the thermal history of the sample as well. Profilometer measurements of the 2.5-μm-thick a-axis film showed surface roughness of about 0.5 μm, while the 1-μm-thick c-axis film has 0.1 μm roughness.

We measured the surface impedance using a cavity resonance technique. We mounted the sample as the end plate of a cylindrical cavity and measured the power as a function of frequency that was transmitted through the cavity for the TE_{011} mode, obtaining the resonant frequency ω_s, and bandwidth $\Delta\omega_s$. We repeated the procedure with a polished oxygen-free high-conductivity (OFHC) copper endplate. The difference between the sample bandwidth and the copper bandwidth is proportional to the difference between the surface resistances of the sample and the copper endplate: $R_s - R_{Cu} = (1/\gamma)(\Delta\omega_s - \Delta\omega_{Cu})/(\omega_{Cu})$, where γ is the resonator constant for the endplate of the cavity. For the 100 GHz cavity, our surface resistance sensitivity is ± 4 mΩ, with absolute uncertainties from run to run of $\Delta R \approx \pm 10$ mΩ. For our 150-GHz measurements the absolute uncertainties in frequency give $\Delta R \approx \pm 100$ mΩ.

Since the films are not single crystals and the currents in the TE_{011} mode flow in a circular pattern on the endplates, in this experiment we do not probe the films in a single crystallographic direction. This is especially true for the a-axis film, in which some of the currents in the film flow parallel to the Cu–O planes (the stronger superconducting direction, according to critical current density measurements) and some flow perpendicular to the planes (the weak direction). The experiment on the c-axis film is a fairer measurement of the superconducting properties in a particular orientation, parallel to the Cu–O planes.

In Fig. 1, we show the temperature dependence of the surface resistance with respect to copper at 102 GHz from 5–100 K for the a-axis and c-axis oriented films. The temperature variation of the copper reference is negligible over the range of this plot. In both films, the surface resistance at 5 K has dropped by close to a factor of nearly 100 from its average value above the transition temperature.

The oscillations apparent above 60 K are due to the fact that the normal-state skin depth is comparable to the thickness of the films, and some power leaks through the film, forming standing waves in the substrate. The oscillations as a function of temperature are produced by the strong temperature dependence of the dielectric constant of $SrTiO_3$, which rises from about 1000 at 100 K to 25 000 at 4 K.[8] Essentially, there are two sources of loss that contribute to the measured surface resistance, resistive losses in the film and losses due to transmission through the film and out of the cavity.

Using the data and a transmission line model for the

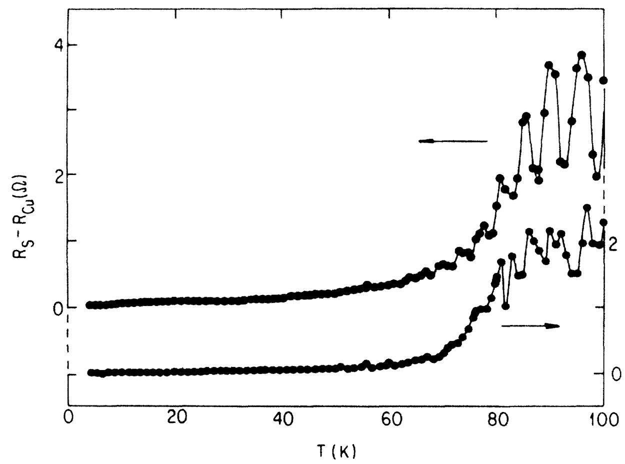

FIG. 1. The surface resistance at 102 GHz, measured with respect to copper as a function of temperature $R_s(T) - R_{Cu}(T)$ for two films with $T_c \approx 85$ K. The top curve is for the a-axis oriented film and the bottom curve (with the vertical scale displaced downward by 1 Ω) is the c-axis oriented film.

transmissive and resistive losses, we estimate the magnitude of the normal-state surface resistance of the film alone, R_n, to be 2.0 Ω [implying a normal-state conductivity σ_n of 1000 $(\Omega\,\mathrm{cm})^{-1}$ and a normal-state skin depth, δ_n (100 GHz), of 5 μm] for the a-axis film, and 1.0 Ω [$\sigma_n \approx 250 (\Omega\,\mathrm{cm})^{-1}$; δ_n (100 GHz) ≈ 2.5 μm] for the c-axis film, both conductivity values being in reasonable agreement with dc measurements on similar films.[4]

In Fig. 2, we have plotted the surface resistance of the films with respect to copper, as in Fig. 1 but on expanded

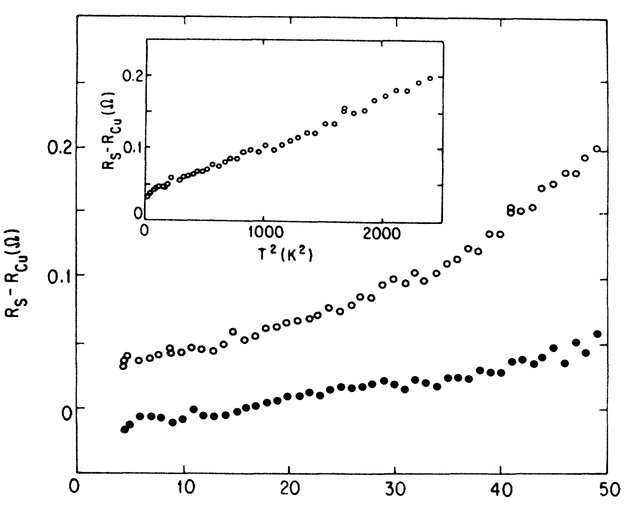

FIG. 2. The surface resistance at 102 GHz, measured with respect to copper, as a function of temperature below 50 K for the a-axis oriented film (top curve) and the c-axis oriented film (bottom curve). Inset: The surface resistance at 102 GHz, measured with respect to copper $R_s - R_{Cu}$, as a function of T^2 below 50 K for the a-axis oriented film.

temperature and resistance scales. On this graph, the temperature variation of the copper reference is negligible. The oscillations are not apparent, even on the expanded scale, below 50 K for both films, which implies that the leakage through the films is indeed small at low temperatures. The surface resistance for the c-axis film becomes lower than our copper endplate below 20 K, which leads to the negative values with respect to copper below 20 ± 10 K. At low temperatures, the measured surface resistance does not saturate to a low, constant value as would be expected from an isotropic BCS superconductor with a thermally activated temperature dependence of σ_1.[3] Instead, we find a measurable dependence all the way down to 5 K. What seems most significant is that the temperature dependences differ between the two orientations: $R_S(a \text{ axis}) \propto T^2$ while $R_S(c \text{ axis}) \propto T$. The quadratic temperature dependence for the a-axis film is brought out in the inset to Fig. 2: $R_s(T^2)$.

Well below the transition temperature, the reactive response dominates the electromagnetic properties. Essentially, as the temperature is decreased, σ_2 increases rapidly, the penetration depth decreases, and both resistive and leakage losses should drop. The transmission line model gives the leakage contribution to the measured surface resistance as

$$R_s(\text{leakage}) = 4\,(\omega\mu_0\lambda)^2/\eta_{\text{eff}}\exp(-2t/\lambda)\ ,$$

where t is the thickness of the film and η_{eff} is the effective impedance of the substrate.

Two sets of muon spin-relaxation measurements have been reported[9,10] that agree on a value for the low-temperature penetration depth, $\lambda(T=0\text{ K}) \approx 1300$ Å. These measurements find that a temperature dependence for λ is consistent with BCS theory. Measurements from magnetization data on single crystals yield estimates for the value of λ ($T=0$ K) of 1800 and 270 Å for currents along the c axis and perpendicular to the c axis, respectively.[11] Quite independently of the spread in values for λ, these results imply that below 50 K the losses due to transmission through our films should be immeasurably small, which agrees with our observation that the oscillations in R_s are not present at low temperatures within our experimental resolution. We are thus led to the conclusion that the observed residual losses arise in the sample itself. It is difficult to say definitively whether they are intrinsic or extrinsic to the superconductor, however.

If we assume that all of the losses in the film result from resistive losses as described by Eq. (2), then a determination of the ratio σ_1/σ_2 depends on the value of λ, which we do not directly measure. As an example, choosing λ $(T=0)=1300$ Å (Refs. 11 and 12) and $R_s(T)=10$ mΩ could give $\sigma_1 \approx \sigma_2/5$ [$(\gg \sigma_n(100\text{ K})$] at low temperatures for the c-axis film, which is totally inconsistent with BCS theory.

A nonvanishing σ_1 at 100 GHz at low temperatures could indicate a constant single-particle density of states as in a gapless superconductor,[12] or the existence of points or lines of nodes in the gap function as in a non-s-wave superconductor. These intrinsic mechanisms should also produce a non-BCS temperature dependence of the penetration depth with a linear or quadratic variation with temperature, as has been suggested for heavy-electron superconductors[13] or $YBa_2Cu_3O_{7-\delta}$ powders.[14] As mentioned above, however, muon spin-relaxation measurements tend to support the BCS theory's predictions, although the error bars in these measurements do not absolutely rule out a somewhat stronger temperature dependence at low temperatures.[14]

Nonintrinsic loss mechanisms could arise from normal surface layers, normal inclusions, or grain-boundary effects. Losses due to grain-boundary effects presumably depend on the microscopic structure of the films, about which too little is known at present to model them usefully. Surface resistance measurements on smoother, more highly oriented films with cleaner surfaces will be necessary to clarify the precise origin of the low-temperature losses in these films.

To make an initial evaluation of the frequency dependence of the surface resistance, we also measured the c-axis film at 148 GHz. Our results at low temperatures are shown in Fig. 3 compared with our 102-GHz data. Despite the higher noise level in the higher frequency measurement, a similar linear temperature dependence appears at low temperatures. Least-squares fits $[R_s/(T) - R_{\text{Cu}}(T) = A + BT]$ for the two sets of data below 50 K gives A (102 GHz) $= -0.021$ Ω, B (102 GHz) $= 0.0013$ Ω/K and A (148 GHz) $= 0.090$ Ω, B (148 GHz) $= 0.0030 \pm 0.0003$ Ω/K. These fits are represented by the straight lines in Fig. 3. The ratio of the slope of the temperature dependences: $(0.0030\ \Omega/\text{K})/(0.0013\ \Omega/\text{K}) = 2.3 \pm 0.3$. Assuming a power-law frequency dependence $R_s \propto \omega^a$, then gives $a \approx \ln(2.3)/\ln(148/102) \approx 2.2 \pm 0.3$. This result agrees reasonably well with the predicted frequency dependence for resistive losses in a superconductor, $a \approx 2$.

In conclusion, we have made some initial measurements

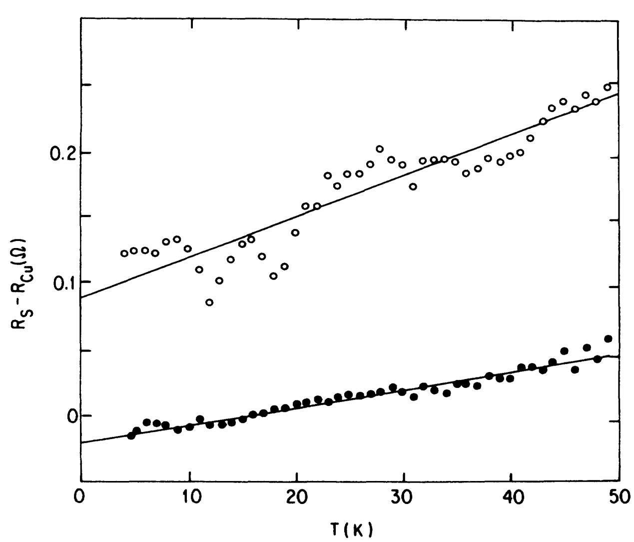

FIG. 3. The surface resistance at 102 GHz (bottom curve) and 148 GHz (top curve), both measured with respect to copper, as a function of temperature below 50 K for the c-axis oriented film. The straight lines represent the linear fits described in the text.

of the millimeter-wave surface resistance in polycrystalline thin-film samples with reasonably good low-frequency superconducting properties. We find clear evidence of a difference in the temperature dependence of the surface resistance between films with different grain orientations. Whether these differences are strictly due to anisotropy of the superconducting properties obviously cannot yet be answered. At 4.2 K, an upper limit of 15 ± 10 mΩ for the residual surface losses at 100 GHz has been established. At temperatures below $T_c/2$, the temperature dependence and magnitude of the losses are consistent with BCS theory, but the $\omega^{2.2}$ dependence of the loss for the c-axis film is consistent with the quadratic frequency dependence characteristic of a superconductor.

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