SURFACE IMPEDANCE STUDIES OF THE HIGH-T_C OXIDE SUPERCONDUCTORS

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Abstract

We review briefly our results on measurements of the millimeter wave surface impedance of ceramic, thin film and single crystal samples of the high-T_C oxide superconductors. The observed losses and temperature dependences do not agree with BCS theory. We discuss our recent measurements of single crystal $Bi_2CaSr_2Cu_2O_x$ and the effect of the application of a perpendicular magnetic field. We consider the possibilities of intrinsic and defect mechanisms to explain our observations. In particular, we discuss the effects of weakly coupled granular films on surface impedance.

Introduction

The surface impedance of the high-T_C oxide superconductors may provide important clues to the underlying physics in these superconductors and will determine the feasibility of many potential technological applications. Important information concerning the energy gap, the symmetry of the superconducting order parameter, and the conduction anisotropy can be obtained from surface impedance measurements. The very low ac losses which might be obtained using high-T_C superconductors would be important in a variety of applications, particularly those which deal with high frequency, low loss transmission lines. We have measured the surface impedance of several high-temperature oxide superconductors, including $YBa_2Cu_3O_x$, $Y_2Ba_4Cu_8O_x$ and $Bi_2CaSr_2Cu_2O_x$. Depending on the material the samples were in ceramic, polycrystalline or oriented thin films, or single crystal form. In each case our measurements show large residual losses with temperature dependences unlike those expected for BCS superconductors. We cannot so far identify with certainty the source of the losses. We discuss the possibilities of intrinsic losses and those arising from material nonidealities and conclude that, even in the best samples (i.e. highly oriented c-axis thin films), grain boundaries are likely to play a major role in determining residual loss.

Measurement Technique

We have measured the surface impedance of our samples using a cylindrical copper cavity in the TE_{011} mode at 102 GHz and 148 GHz.[1,2,3] The samples form one of the endwalls of the cavity. We use a sample replacement technique in which we exchange the sample endwall with a polished copper endplate and thereby measure the surface impedance with respect to copper. The total surface impedance is described by its real and imaginary parts, $Z_S=R_S+jX_S$. In general we suppose that for the surface resistance, $R_S(T)=R_S(0)+\Delta R_S(T)$, and for the surface reactance, $X_S(T)=X_S(0)+\Delta X_S(T)$. This technique is an effective way to measure $R_S(T)$ and $\Delta X_S(T)$. $X_S(0)$ is difficult to obtain using this technique, though we have had some success for thin films[3] and ceramics[2].

Results

In table 1 we present a summary of our measurements to date. The samples are ordered according to their low temperature surface resistance with the best samples first. For reference we also show the predictions of the Mattis-Bardeen theory. The columns of the table show (reading from left to right) the low temperature surface resistance, R_S, the temperature dependence of the surface resistance, ΔR_S, the temperature for which the surface resistance of the superconductor is equal to that of copper, the low temperature penetration depth, λ, the temperature dependence of the penetration depth, $\Delta\lambda$, and the frequency dependence of the surface resistance.

For all the samples the surface resistances measured are much larger than what the Matthis-Bardeen theory predicts assuming a BCS superconductor with a transition temperature of 90 K. Magnitudes of surface resistance vary greatly from sample to sample, indicating that sample quality is of crucial importance. Our best samples, c-axis thin films, have surface resistances less than copper at 35±10 K at 150 GHz. C-axis films of $YBa_2Cu_3O_x$ and $Y_2Ba_4Cu_8O_x$ show the smallest losses, while a-axis oriented films and ceramics of $YBa_2Cu_3O_x$ show losses more than an order of magnitude larger. The losses in $Bi_2CaSr_2Cu_2O_x$ single crystals have been very large so far and show great sensitivity to the cleaving of the surfaces. The losses will undoubtedly improve dramatically with better crystals and/or better surface preparation. The surface resistance and surface reactance of c-axis films of $YBa_2Cu_3O_x$ and $Y_2Ba_4Cu_8O_x$ are remarkably similar at low temperature both in magnitude and in temperature dependence. C-axis films consistently show $R_S(T)-R_S(0)\sim T$ and $X_S(T)-X_S(0)\sim T^2$ (for a thick film where $X_S>>R_S$, $X_S=\omega\mu_0\lambda$, λ=penetration depth) in contrast to the exponential behavior predicted by Matthis-Bardeen theory. Our measurements[1] of c-axis films are consistent with an ω^2 frequency dependence of the surface resistance as expected for any two fluid type model of a superconductor, but the ceramics[2] show ω^{α} where $1<\alpha<2$. We will comment on this further below.

The surface impedance of c-axis films shows no sensitivity to the application of perpendicular magnetic fields up to 300 Gauss. This is in marked contrast to the extreme sensitivity of the surface losses of ceramic samples to very small applied fields reported by others[4].

For our best c-axis films the superconducting transitions measured by the change in surface impedance are typically 10 K wide and show substantial reduction in the surface resistance as compared to the normal state some 5 or 6 degrees below the typical zero resistance dc transition temperature of 87 K. The typical dc transition width is about 3 K. This is possibly due to the granularity of the films and the distribution of transition temperatures in the grains and grain boundaries. We point out, however, that the this difference between ac and dc transitions is much smaller in the ceramics than in the thin films.

Discussion of the Results

The low temperature behavior described above is much different from the predictions of the BCS theory and show large residual losses at low temperatures. It is important to understand the implications of these results both from the fundamental and practical points of view. Of primary concern is our ability to distinguish effects which are intrinsic to the ideal superconducting material from those which are the result of material inhomogeneities such as grain boundaries, normal inclusions, surface layers, structural defects and imperfect or nonuniform doping. The observed temperature dependences of $R_S(T)-R_S(0) \sim T$ and $X_S(T)-X_S(0) \sim T^2$ could be indications of a superconductor with nodes in the energy gap[5] or other intrinsic effects as we have discussed elsewhere[3]. On the other hand, the large losses in the ceramic materials and their sensitivity to small magnetic fields are almost certainly due to the weak coupling between grains in these tenuously connected materials. We have proposed earlier[6] that the surface impedance in our best c-axis thin films may also be strongly affected by the coupling between the grains, even though these films show surface resistances more than an order of magnitude smaller than the ceramics and do not exhibit great sensitivity to the application of small magnetic fields. Though this model cannot explain on a fundamental level the origin of the observed temperature dependences, it does make clear the possibility that both the surface resistance and the surface reactance may be dominated by the properties of the grain boundary, even in our oriented, *c*-axis thin films. Because of our success with this model we feel that any interpretation of the measurements of the properties of the high-T_C superconductors should seriously consider the effect of grain boundaries.

Single crystal samples of $Bi_2CaSr_2Cu_2O_x$ similar to those discussed above that show large residual RF losses, show, by contrast, much better sandwich[7] and point contact[8] tunneling results than oriented thin films similar to those that show the smallest RF losses. The improved tunneling results are likely related to the particularly clean surfaces obtainable in $Bi_2CaSr_2Cu_2O_x$ as compared with $YBa_2Cu_3O_x$. Considering the sensitivity of the surface resistance to the cleaving of the sample surface, it seems likely that in order to obtain low values of the surface resistance in the single crystals the surfaces will have to be smooth as well as clean. Typical cleaving techniques we have used leave a surface with many visually apparent steps, corresponding to cleavage at planes of different depths across the surface and general surface damage.

Weakly Coupled Grain Model

In reference 6 we developed a simple model of the effect grain boundaries on the surface impedance of our *c*-axis thin films. The film is modeled as a network of ideally superconducting grains coupled by Josephson junctions, which we describe using the standard resistively shunted junction with negligible capacitance. In figure 1 we show the basic element of the superconducting network, a superconducting grain in series with a grain boundary, and the corresponding circuit model of this basic element. The important parameters of the model are the junction I_CR product (I_C is the junction critical current) and the effective grain size *a* . In the figure, $\mathcal{L}_G=\mu_0\lambda_{ab}^2$ is the kinetic inductivity of the grain for current flow in the ab plane (penetration depth λ_{ab}), $\mathcal{L}_J=\hbar/2eJ_C$ is the unit areal inductance of the junction (J_C is the junction critical current density), and $\mathcal{R}$ is the unit areal shunt resistance ($R=\mathcal{R}\times$(junction area)). $\mathcal{L}_J/a$ and $\mathcal{R}/a$ are the effective parallel inductivity and resistivity of the grain boundary averaged over the grain dimension, i.e., in the continuum approximation. We show in reference 6 that the surface impedance resulting from this model is

$$Z_S = \omega \mu_0 \lambda_{eff} \left[j + \frac{1}{2} \left(\frac{\lambda_J}{\lambda_{eff}} \right)^2 \frac{\hbar \omega}{2 e I_C R} \right] \text{, where}$$

$$\lambda_{eff} = \sqrt{\lambda_{ab}^2 + \lambda_J^2} \quad \text{and} \quad \lambda_J = \sqrt{\frac{\hbar}{a\, 2\, e\, J_C\, \mu_0}} .$$

We point out that λ_J is *not* the Josephson penetration depth, though the form of the equation above is very similar to that of the equation for the Josepson penetration depth. The expression for the surface impedance above is valid in the limit $R_S \ll X_S$, which is the relevant limit for our c-axis films. Applying this model to our measurements of the surface impedance of c-axis films yielded a =0.4 μ and $I_C R$=1.5 mV at low temperature, which is in reasonable agreement with our knowledge of thin film morphology and other measurements of the properties of grain boundaries[9].

The model above shows $R_S \sim \omega^2$, and our measurements are consistent with this frequency dependence[1]. If the capacitance in the junction had been significant, then the grain boundary model would not have predicted *an ω^2 dependence. Hence, our neglect of the capacitance in the grain* boundary is consistent with our observations. A rough estimate of the capacitance of a grain boundary in a c-axis film further substantiates the negligible effect it will have on the surface impedance at these frequencies. If we add a parallel capacitance per unit area, $C_J \approx \varepsilon/t$ (t=junction thickness) to the circuit of figure 1, then this capacitance will have a negligible effect as long as the conduction through the junction is dominated by the inductive channel. This is true if $\omega \mathcal{L}_J << 1/\omega C_J$ or $\omega^2 << \omega_p^2$, where $\omega_p = (1/\mathcal{L}_J C_J)^{1/2}$ is the junction plasma frequency. If we suppose that $t \approx 10$Å and $\varepsilon \approx 10\varepsilon_0$ (typically $J_C = 5 \times 10^6$ A/cm^2) then $\omega_p \approx 4 \times 10^{13}$ sec^{-1}. Since $\omega \approx 10^{12}$ sec^{-1} (150 GHz) the capacitance is almost certainly negligible in our c-axis films.

The grain boundary model above for c-axis films is likely to be an inadequate description of the effects of grain boundaries in ceramic materials. All measurements of the properties of the ceramics indicate that the grains are much more weakly connected than the grains of high quality c-axis thin films. In addition, the randomly oriented nature of the grains in the ceramics is bound to produce percolation effects not present in the films because of the anisotropic conduction properties of these materials. Hence, it is not at all surprising that significant differences exist in the surface impedances of the ceramics and the thin films. In particular, the observation of a frequency dependence other than ω^2 in the surface resistance of ceramic materials is not unreasonable. Even in the grain boundary model of the thin film surface resistance frequency dependence other than ω^2 can be obtained by letting the conduction in the resistive or capacitive channels become comparable to the conduction in the inductive channel.

The grain boundaries must also contribute to the normal state resistivity of the film. We can use our knowledge of the critical current density of the film to estimate the effect of R on the normal state resistivity,

$$\rho_{eff} = \rho_{grain} + I_C R / J_C a,$$

where ρ_{eff} is the effective normal state resistivity including the effects of the grains and the grain boundaries and ρ_{grain} is the intrinsic normal state resistivity of the grain. Substituting in the values listed above yields $\Delta\rho = \rho_{eff} - \rho_{grain} = 7.5\ \mu\Omega\text{-cm}$. Typically we measure $\rho_{eff} \approx 100\ \mu\Omega\text{-cm}$ just above the transition in $YBa_2Cu_3O_x$ films so that the grain boundaries add only a small amount to the overall resistivity. We have assumed in the above that R is not a function of temperature by extrapolating the low temperature value to temperatures above the transition. This is almost certainly not the case. Our data imply that R should decrease with temperature, indicating that the effect of the grain boundaries on the total resistivity in the normal state is even smaller than that stated above. Hence, although the grain boundaries may dominate the RF losses in the superconducting state, they probably have negligible effect on the properties of the normal state. This implies that a film with very good normal state properties (such as resistivity versus temperature) may still have large RF losses.

It is also interesting to consider the real part of the conductivity in the grain boundary due to the resistive channel. If t is the thickness of the grain boundary then

$$1/\sigma_{GB} = \rho_{GB} = I_C R / J_C t \approx 3000\ \mu\Omega\text{-cm}$$

where σ_{GB} is the real part of the conductivity in the junction region and we have assumed for the sake of argument that t=10Å. Though we cannot comment definitively on the meaning of this number, we point out that the resistivity shown above is comparable to the normal state resistivities we measure in poor quality films. Such films often include large amounts of secondary phases (especially when the stochiometric composition of the film is incorrect). Hence it is not inconceivable that the grain boundaries consist of second phase material or poorly doped intrinsic material. These are, of course, not the only candidates for the creation of a weak link.

Conclusion

Better samples are necessary to resolve the issue of the source of the large values of the surface impedance we have observed thus far. We remain cautiously optimistic about the potential applications of these superconductors in low loss transmission lines, given the trend toward decreasing surface resistance with improved sample quality and the possibility that all of our observations to date may be dominated by grain boundary effects. Better quality thin films will require the growth of the superconducting material in-situ, as opposed to the amorphous deposition, post annealing technique we have used thus far.

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Table 1. Observed Properties of the Surface Impedance of High-T_C Oxide Superconductors

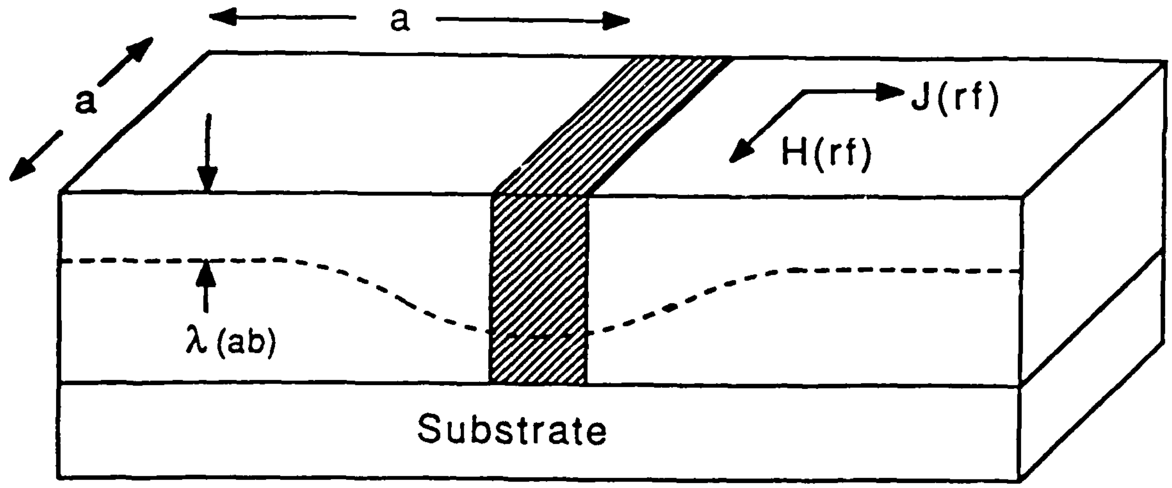
	Surface Resistance R_S at 4.2 K (Ω)	$\Delta R_S(T)$ ~? ($T \ll T_C$)	T for $R_S=R_{Cu}$† (K)	Penetration Depth, λ††, at 4.2 K (Å)	$\Delta\lambda(T)$ ~? ($T \ll T_C$)	$R_S(\omega)$ ~? ($T \ll T_C$)
$YBa_2Cu_3O_x$ c-axis film 148 GHz[1,3]	0.015±0.01	T	35±10	3000±1000	T^2	ω^2
$Y_2Ba_4Cu_8O_x$ c-axis film 148 GHz[3]	0.015±0.01	T	35±10	3000±1000	T^2	---
$YBa_2Cu_3O_x$ a-axis film 102 GHz[1]	0.3±0.01	T^2	$R_S>R_{Cu}$ for all T	---	---	---
$YBa_2Cu_3O_x$ ceramics 102 GHz[2]	0.65±0.01	$T^{1+\varepsilon}$ $0<\varepsilon<1$	$R_S>R_{Cu}$ for all T	9000±3000	$T^{1+\varepsilon}$ $0<\varepsilon<1$	ω^α $1<\alpha<2$
$Bi_2CaSr_2Cu_2O_x$ single crystal ab plane 150 GHz	0.8±0.01	T	$R_S>R_{Cu}$ for all T	---	T^2	---
Mattis-Bardeen and BCS, 100 GHz T_C=90K	$<10^{-6}$ (25 K)	$e^{-\Delta/kT}$	---	640†††	$e^{-\Delta/kT}$	ω^2

†$R_{Cu} \approx 40$ mΩ at 102 GHz, T<50 K
≈ 55 mΩ at 148 GHz, T<50 K

††$\lambda = X_S/\omega\mu_0$ = reactive skin depth = penetration depth for $R_S \ll X_S$

†††$\lambda = c/\omega_{plasma}$, $\omega_{plasma} \approx 3.1$ eV, valid in the clean limit only

a



b

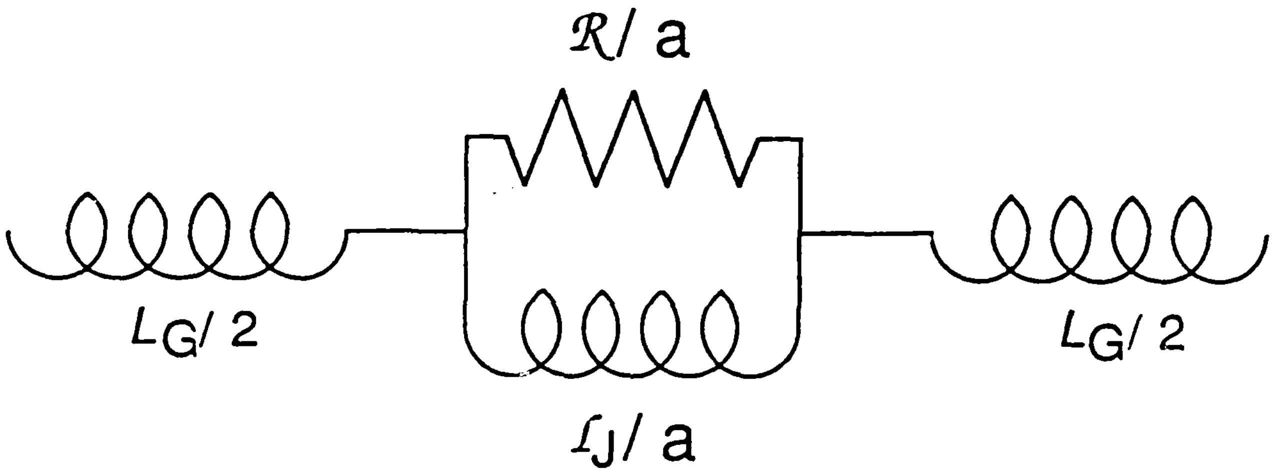


Figure 1a. Two grains of dimension a are coupled by a weakly superconducting region (shaded) in this idealization of a grain boundary junction of a thin film. The currents are confined to a penetration depth as in a typical surface impedance measurement. The dashed curve is a profile of the penetration depth of the currents into the grain and grain boundary. b. A circuit model of figure 1a consists of purely inductive grains in series with a grain boundary of parallel inductive and resistive channels.