# MICROWAVE PROPERTIES OF HIGH TRANSITION TEMPERATURE SUPERCONDUCTING THIN FILMS



**Final Technical Report to the NASA Lewis Research Center**

**Contract #NCC 3-40**

Dr. W. L. Gordon
Department of Physics
Case Western Reserve University
Cleveland OH. 44106

June 21, 1991

# TABLE OF CONTENTS

# I. INTRODUCTION

The discovery oh high temperature supeconductors (HTS) has prompted efforts to develop their microwave applications [1,2]. The low microwave and millimeter wave losses of these new superconducting copper oxides make them very attractive for the development of voltage dividers, resonators, phase shifters, filters, and other high frequency analog devices. Millimeter and microwave characterization of HTS materials have been performed through using different experimental techniques [3-5]. Among these, are studies related to the response of the HTS compounds to electromagnetic radiation. The study of the electromagnetic response allows the determination of material parameters such as the magnetic penetration depth ($\lambda$), the complex conductivity ($\sigma_* = \sigma_1 - j\sigma_2$), and the surface impedance ($Z_s = R_s - jX_s$). Knowledge of these parameters is of importance for technological applications since they are directly related to the losses of electromagnetic energy in these materials during signal propagation.

In the past, millimeter and microwave transmission and reflection experiments have been used to study the properties of low transition temperature superconducting (LTS) thin films [6-8]. Results from these experiments provided strong evidence for the validity of the Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity. Therefore, transmission-reflection experiments are not only useful as a convenient characterization mechanism but provide a powerful technique for the analysis of the nature of the superconducting state in the HTS materials.

During the course of this contract, we performed extensive studies of the interaction of microwaves with $YBa_2Cu_3O_{7-\delta}$, Bi-based, and Tl-based superconducting thin films deposited in several microwaves substrates. The data were obtained by measuring the microwave power transmitted through the film in the normal and the superconducting state and by resonant cavity techniques. Our main motivations for this work were to quantify and understand the physical parameters such as $\lambda$, $\sigma^*$, and $Z_s$, of HTS materials at microwave frequencies. Based on these parameters we discuss the suitability of these HTS thin films for microwave applications.

# II. Measurement Apparatus and Procedures

A schematic of the configuration used to measured the microwave power transmitted through the HTS thin films and the microwave substrates analyzed in this study is shown in Fig. 1. The main components of the experimental apparatus are a HP-8510B automatic network analyzer, a closed cycle helium gas refrigerator, and a temperature controller (LakeShore Cryotronics, model DRC 91C), which are controlled by a HP 9000-216 computer. The network analyzer is coupled to the refrigerator by Ka-frequency band (26.5 to 40.0 GHz) rectangular waveguides. The measurement technique is based on comparing the reflected and the transmitted signals against the incident microwave signal produced by a 0.01 to 20 GHz synthesized sweeper whose output is doubled to the Ka-frequency band. Through the use of a directional coupler the incoming signal is divided, with part of it directed toward a mixer and used as a reference signal and the rest of the signal directed toward the sample. A second directional coupler is used to direct a portion of the signal reflected from the sample to a second mixer. The fraction of the original signal which is transmitted through the sample is also directed to a third mixer by means of a directional coupler. A 0.01 to 26.5 GHz synthesized sweeper is used as a local oscillator to

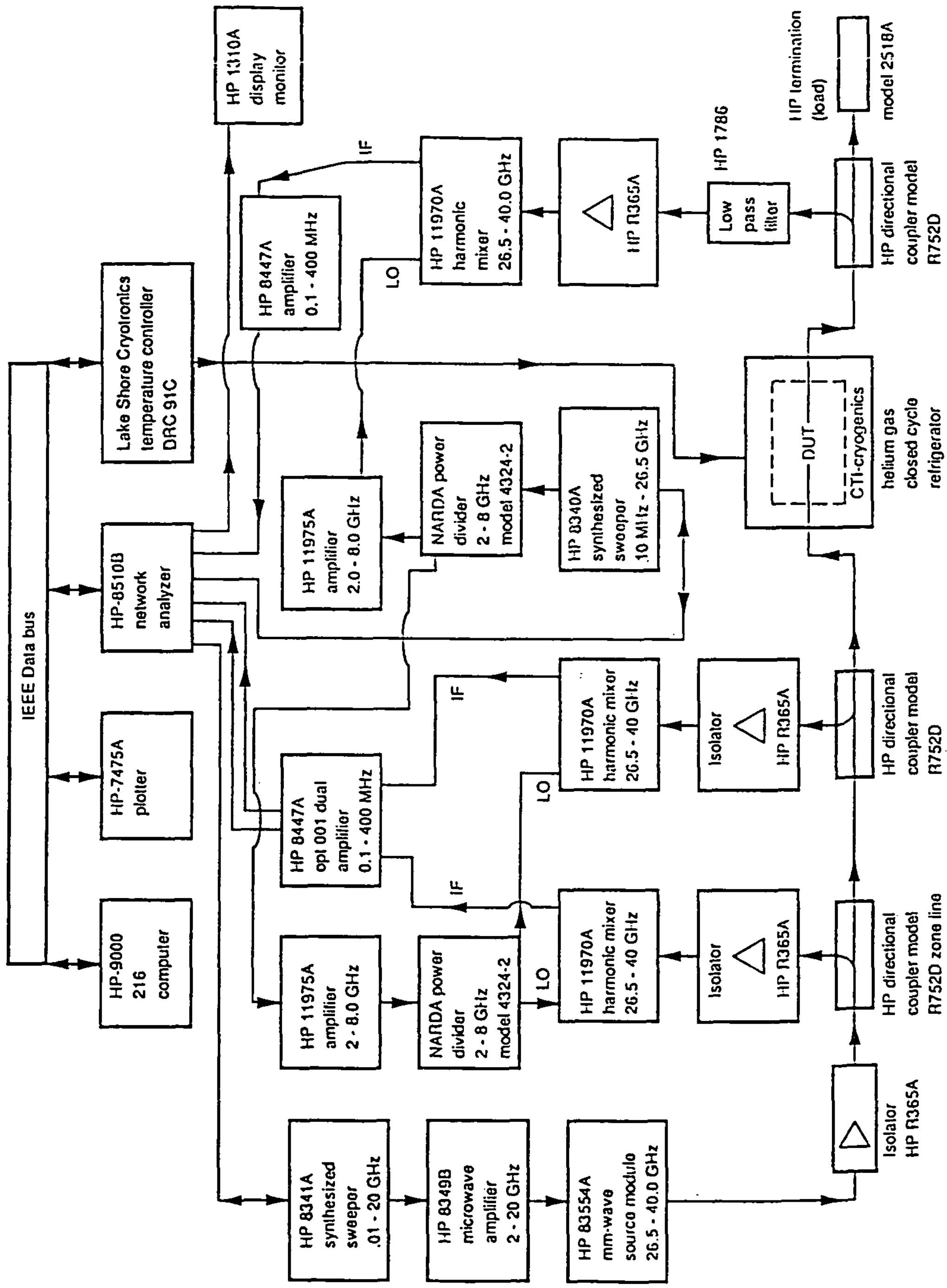


Figure 1: Microwave measurements apparatus.

feed the three mixers. The intermediate frequency signals from these mixers are fed into the network analyzer. The network analyzer compares the power and phase of the reflected and transmitted signals with that of the reference signal to determine the reflection and transmission coefficients. The data measured in this way are then stored by the computer for subsequent analysis.

All the measurements were performed under vacuum (usually less than 10 mtorr), in a custom made aluminum vacuum chamber designed to fit on top of the external shield of the refrigerator and to give access to the set up of waveguides connecting the network analyzer with the refrigerator. In order to preserve the vacuum, natural mica windows were used at the coupling points between the external waveguides and the waveguides inside the chamber. Mica was selected for the windows because of its very low loss, thermally stable relative dielectric constant, and its transparency to microwave signals in the temperature and frequency ranges considered in this study. Inside the vacuum chamber the sample was oriented perpendicular to the microwave source by clamping it between two waveguide flanges thermally connected to the cold head of the refrigerator through a copper plate. A thin layer of In was used between the holding flange and the film in order to improve both thermal and electrical contact between them. For HTS measurements, the film side of the sample was directed toward the incident microwave signal, as shown by the schematic in Fig. 2. The system was calibrated before the beginning of each measurement cycle. The calibration was performed using the Hewlett Packard WR-28 Calibration Standards Kit R11644A consisting of a thru, a short circuit, a shielded open circuit, and loads which provides corrected transmission and reflection measurements. According to Hewlett Packard [9], the measurement repeatability (sweep-to-sweep) using a through or short

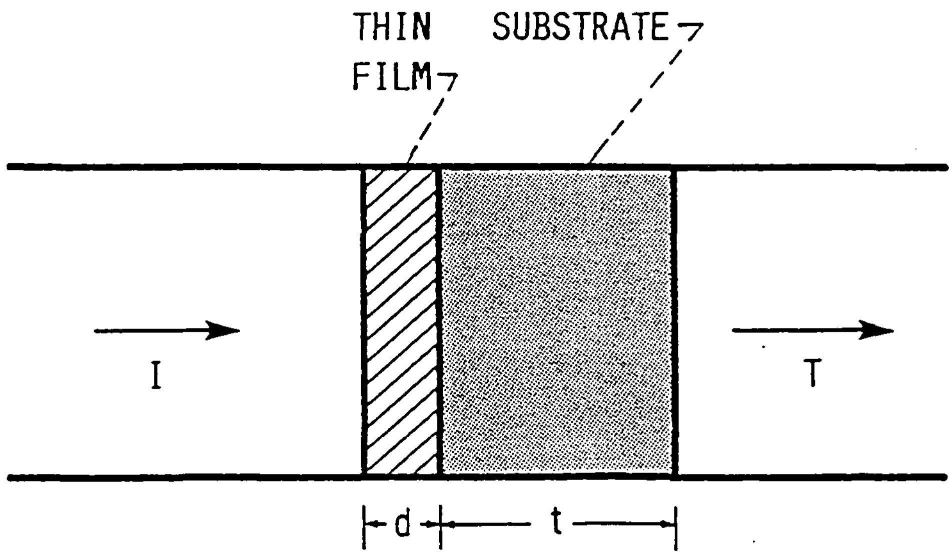


Figure 2: Side view of a rectangular waveguide propagating the $TE_{01}$ mode with its entire cross section covered by a high-$T_c$ superconducting thin film of thickness d deposited on a dielectric substrate of thickness t and refractive index n. In this work I ~ 16 mW/cm$^2$, d ~ 2000 Å, t ~ 5.0 x $10^{-2}$ cm, and $10^{-6} \leq \mathbf{T/I} \leq 10^{-1}$.

as the device under test is within 0.005 dB for the magnitude and 0.5 degrees for the phase of the transmitted and reflected signals.

The noise level for our measurements was determined to be below -60 dB after the calibration. Corrections for background attenuation and phase, obtained by measuring the transmitted power as a function of temperature in the absence of the sample, were made by subtracting this contribution from the data obtained with the sample in place. Measurements made with the bare substrates were used to determine any temperature dependence of their dielectric constant.

The microwave power transmission coefficient T was measured with a repeatability of ± 5% over the entire temperature range. This error was estimated by measuring several samples repeatedly, with the samples being removed and replaced between each set of measurements. A run started at room temperature, with measurements taken during cooling. The temperature of the film was measured by two silicon diodes (LakeShore Cryotronics, model DT-470-LR-13), which were placed in a 1/8 inch diameter hole on top of each of the sample's supporting flanges. The accuracy of these diodes is ± 1.0 K from 1.4 to 100 K, and 1% of the actual temperature in the range from 100 to 325 K. The difference in the temperature readings of the two sensors was less than 0.2 K over the entire temperature range. An additional sensor was located next to the heater on the cold head of the refrigerator and was used to control the temperature of the cold head.

To determine if there were any effects due to a thermal gradient between the HTS sample and the temperature sensors, the temperature dependence of the power transmission coefficient was measured for a few samples during cooling and also while warming to temperatures above $T_c^{mw}$. It was found that the maximum difference in the measured power at a given temperature for the two measurements was less than 6%, and typically was less than 3%. This difference was in the sense expected from a temperature gradient, but can be neglected for the purposes of this study.

To investigate any possible warming of the HTS sample as a result of the incident microwave power (~ 16 mW), power transmission measurements were performed at other incident power levels (1.0 mW and 0.1 mW). Since no shift of the $T_c^{mw}$ within a resolution of 0.5 K was observed for the different power levels employed, we concluded that any change in the temperature of the film due to absorbed microwave power can be neglected.

# III. Superconducting Films and Microwave Dielectric Substrates

In this study we concentrated our efforts in the study of thin films of the three principal HTS discovered so far; $YBa_2Cu_3O_{7-\delta}$, Bi-Sr-Ca-Cu-O, and Tl-Ba-Ca-Cu-O. These films which ranged in thickness from ~ 800 Å to ~ 7000 Å, were deposited by others using laser ablation, co-evaporation, magnetron sputtering, and sequentially evaporation techniques. The films were deposited on several dielectric substrates such as $LaAlO_3$, MgO, YSZ, $LaGaO_3$, and $SrTiO_3$. Some of these substrates were characterized performing microwave signal transmission and reflection measurements in order to determine their dielectric constant ($\varepsilon_r$). These measurements were important since the successful application of thin films made with the new HTS compounds in the development of microwave circuits rest considerably on the dielectric properties of the different substrates used for film deposition. For microwave applications it is desirable to have substrates with low $\varepsilon_r$ and loss tangent ($\tan\delta$) if good performance from microwave components is expected. Results of our measurements on these dielectric substrates were summarized and published in two scientific journals (see publications list, section VI).

# IV. Results and Discussion

We have studied the microwave response of $YBa_2Cu_3O_{7-\delta}$, Bi-based, and Tl-based HTS thin films. We have measured the transmitted power as a function of temperature, incident power, and film thickness. We found that for the three types of HTS materials the microwave transmission properties are weakly dependent on temperature in the normal state, but change drastically upon transition to the superconducting state. In particular, the transmission decreases and there is a negative relative phase shift with respect to the phase at room temperature when the sample is cooled through its transition temperature, as shown in Figs. 3 and 4, respectively.

The magnetic penetration depth for all films was determined from the surface reactance of the films. We found that the smallest penetration depth values were obtained for the $YBa_2Cu_3O_{7-\delta}$ films, whose values of $\lambda$ were consistent with the best values reported by others so far. We were able to verify experimentally the intrinsic anisotropy of $\lambda$ for this HTS superconductor by measuring this parameter in c-axis and predominantly a-axis oriented thin films. As expected from the intrinsic anisotropy of these HTS materials, the value of $\lambda$ for the a-axis oriented film

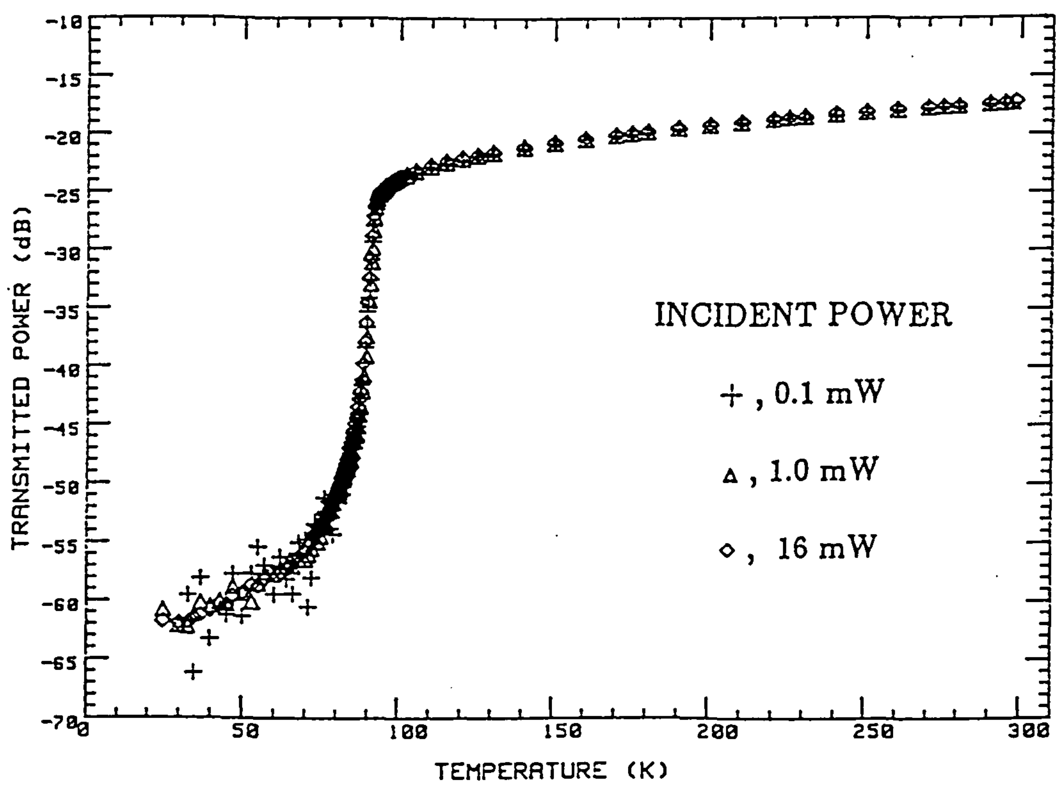


Figure 3: Transmitted power versus temperature for a laser ablated $YBa_2Cu_3O_{7-\delta}$ thin film (2400 Å) on $LaAlO_3$.

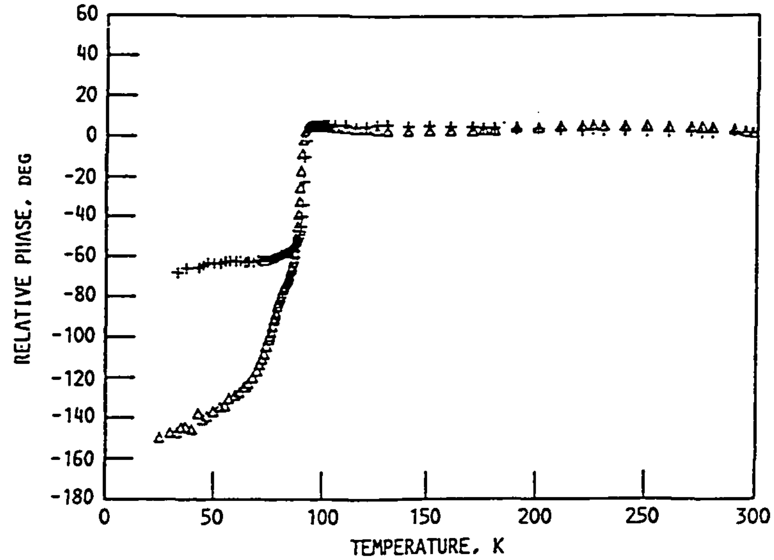


Figure 4: Measured relative phase shift for an off-axis magnetron sputtered $YBa_2Cu_3O_{7-\delta}$ thin film (800 Å) on YSZ (+), and for a laser ablated $YBa_2Cu_3O_{7-\delta}$ thin film (4900 Å) on $LaAlO_3$ (Δ).

was larger than the values obtained for c-axis oriented films. In fact, the ratio of λ for an a-axis oriented film to that of c-axis oriented films agrees very well with that reported by others for single crystals of the $YBa_2Cu_3O_{7-\delta}$ superconductor. We also observed that λ increased with increasing film thickness, which is consistent with the increase of the number of a-axis oriented grains and other structural and material defects with increasing film thickness. From the thickness dependence of λ we were able to determine, for the first time, an intrinsic value of λ for the $YBa_2Cu_3O_{7-\delta}$ superconductor. This value is consistent with that expected from measurements of λ in single crystals by other techniques. Values of λ for $YBa_2Cu_3O_{7-\delta}$ thin films on $LaAlO_3$ are given in table I, while Fig. 5 shows a plot of λ versus film thickness for $YBa_2Cu_3O_{7-\delta}$ thin films.

## TABLE I

MAGNETIC PENETRATION DEPTH λ OF $YBa_2Cu_3O_{7-\delta}$ THIN FILMS ON $LaAlO_3$

| Sample no. | Deposition Method | d (Å) | λ (30 K, nm) |
|---|---|---|---|
| 1 | laser ablation | 4900 | 250 |
| 3 | laser ablation | 2400 | 160 |
| 4 | laser ablation | 1769 | 170 |
| 5 | laser ablation | 828 | 150 |
| 6 | laser ablation | 1762 | 610 |
| 9 | laser ablation | 1000 | 120 |
| 11 | dc mag. sputt. | 2600 | 180 |
| 12 | laser ablation | 2665 | 140 |
| 13 | laser ablation | 2655 | 170 |
| 14 | laser ablation | 4000 | 260 |

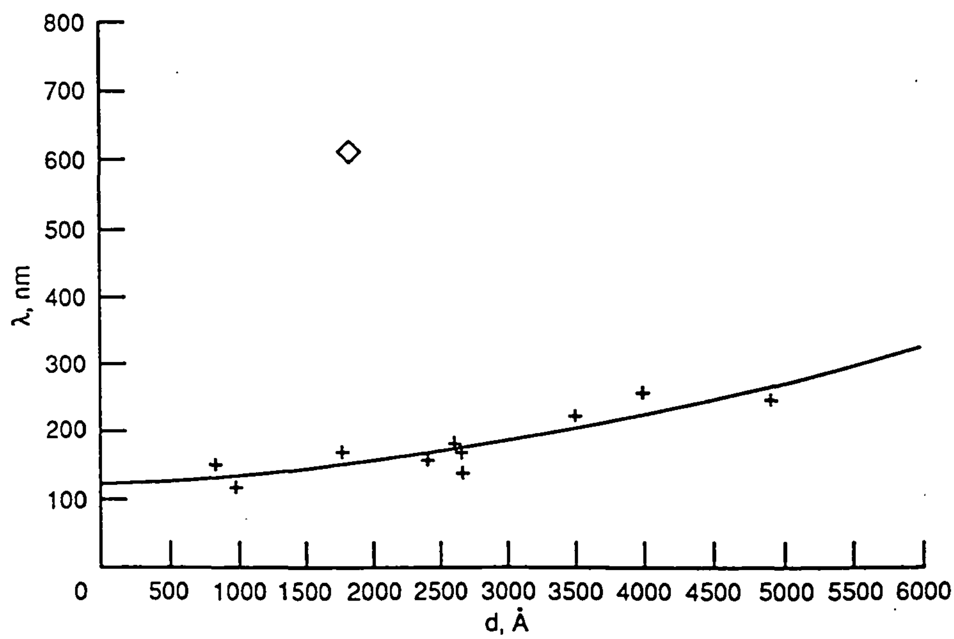


Figure 5: Magnetic penetration depth (λ) versus film thickness (d) for $YBa_2Cu_3O_{7-\delta}$ thin films. The ◊ represents an a-axis oriented film and the solid line represents s second degree polynomial fit to the data.

The $\lambda$ values for the Bi-based films and Tl-based films were larger than those obtained for the $YBa_2Cu_3O_{7-\delta}$ films, and for the Tl-based films the calculated $\lambda$ values were larger than the best reported values to date. We have seen that these large values may be associated with the poor grain connectivity exhibited by these films. Values for $\lambda$ were also determined from the measured $\sigma_2$. It was found that the values of $\lambda$ calculated in this manner were larger than those obtained from $X_s$.

The microwave complex conductivity was determined in both the normal and the superconducting state. The largest values for the conductivity in the normal state were obtained for the $YBa_2Cu_3O_{7-\delta}$. For the three types of HTS films we observed that both $\sigma_1$ and $\sigma_2$ increased upon transition to the superconducting state as shown in Fig. 6. This implies that the temperature dependence of $\sigma_1$ deviates from the predictions of the two-fluid model. In addition, we found that the temperature dependence of $\sigma_1$ is not consistent with that expected from the Mattis-Bardeen equations, and the BCS theory. The largest values for $\sigma_2$ were measured for the $YBa_2Cu_3O_{7-\delta}$ thin films. We found that for the Bi-based and Tl-based films we were able to measure $\sigma_1$ for all measurable temperatures below $T_c^{mw}$, while for most of the $YBa_2Cu_3O_{7-\delta}$ films we found that once the complex conductivity became highly reactive, measurement of $\sigma_1$ turned out to be very difficult for temperatures far below $T_c^{mw}$.

We have calculated the surface resistance $R_s$ for the three types of HTS films studied. We found that for high quality $YBa_2Cu_3O_{7-\delta}$ thin films the $R_s$ values at 77 K compared fairly well with those reported by other researchers for similar films and were equal or smaller than that for copper at the same frequency and temperature (see table II). However, for the Bi-based and Tl-based films the calculated $R_s$ values were larger than those of $YBa_2Cu_3O_{7-\delta}$ thin films and of copper for all the temperatures considered in this study. We have also fabricated a cylindrical

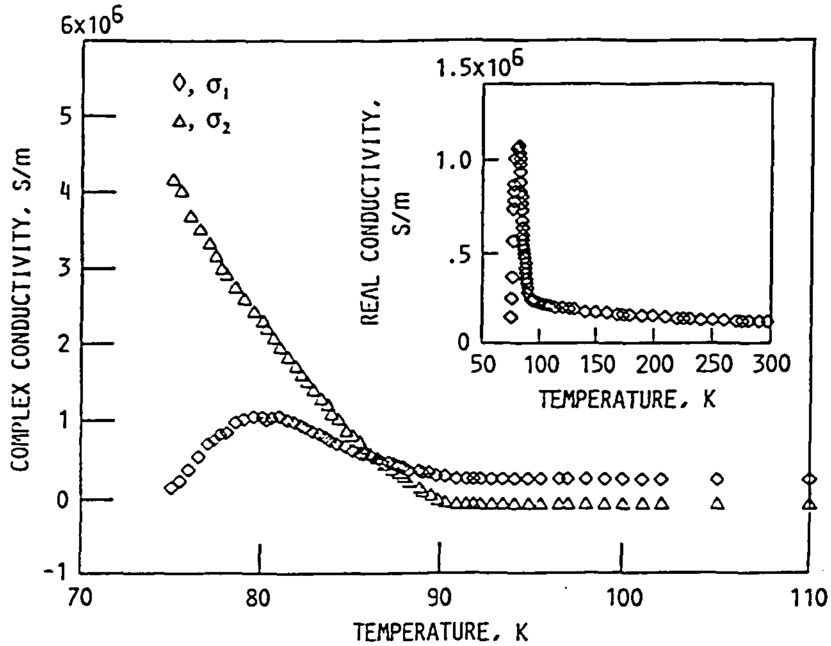


Figure 6: Real and imaginary parts of the microwave conductivity $\sigma^* = \sigma_1 - j\sigma_2$ versus temperature at 30.6 GHz for a laser ablated $YBa_2Cu_3O_{7-\delta}$ thin film (4900 Å, $T_c^{MW}$=90.8 K) on $LaAlO_3$.

copper cavity to measure $R_s$. The measured $R_s$ values for $YBa_2Cu_3O_{7-\delta}$ thin films on $SrTiO_3$ and $LaGaO_3$ were in good agreement with $R_s$ values reported by others using resonant cavity techniques and with those obtained using our power transmission measurement technique, if a quadratic frequency dependence for $R_s$ is assumed.

Our analysis suggests that, among those studied, the laser ablated and dc off-axis magnetron sputtered $YBa_2Cu_3O_{7-\delta}$ thin films are the most promising for microwave applications. However, we believe that improvements in the superconducting properties of Bi-based and Tl-based HTS

TABLE II

Microwave Measured Parameters of YBCO Superconducting Films

| S.N. | SUBSTRATE. | D.M. | d(Å) | $T_c^{mw}$(K) | $R_N^{mw}$(Ω) | $\rho_N^{mw}$(μΩ-cm) | $\delta_N^{mw}$(μm) | ℓ (Å) | $R_S^{eff}$(mΩ) |
|---|---|---|---|---|---|---|---|---|---|
| 1 | $LaAlO_3$ | LA | 4900 | 90.8 | 0.68 | 379 | 5.57 | 7.8 | 29 @77 |
| 2 | YSZ | OAMS | 800 | 92 | 0.37 | 111 | 3.02 | 26.5 | 45 @77 |
| 3 | $LaAlO_3$ | LA | 2400 | 91.6 | 0.51 | 216 | 4.24 | 13.6 | 3.3 @80 |
| 4 | $LaAlO_3$ | LA | 1769 | 91.7 | 0.58 | 274 | 4.72 | 10.7 | 32 @77 |
| 5 | $LaAlO_3$ | LA | 828 | 92.5 | 0.61 | 313 | 5.12 | 9.4 | 112 @77 |
| 6 | $LaAlO_3$ | LA | 1762 | 94.0 | 0.96 | 758 | 7.89 | 3.9 | 566 @77 |
| 7 | MgO | OAMS | 1000 | 94.0 | 0.67 | 373 | 5.57 | 7.9 | 15.3@77 |
| 8 | $LaGaO_3$ | LA | 4000 | 94.0 | 0.95 | 752 | 7.91 | 3.9 | 13 @77 |
| 9 | $LaAlO_3$ | LA | 1000 | 88.6 | 0.45 | 164 | 3.66 | 17.9 | 43 @77 |
| 10 | MgO | LA | 3500 | 93.2 | 0.70 | 408 | 5.83 | 7.2 | 86 @77 |
| 11 | $LaAlO_3$ | OAMS | 2600 | 93.6 | 0.52 | 222 | 4.26 | 13.2 | 6.9 @77 |
| 12 | $LaAlO_3$ | LA | 2665 | 88.8 | 0.60 | 294 | 4.97 | 10.0 | 47 @77 |
| 13 | $LaAlO_3$ | LA | 2655 | 91.2 | 0.70 | 400 | 5.79 | 7.4 | 29 @78 |
| 14 | $LaAlO_3$ | LA | 2655 | 91.4 | 0.80 | 524 | 6.62 | 5.6 | 132 @77 |

S.N.= Sample number, D.M.= Deposition Method, LA= Laser Ablated , OAMS= Off-axis Magnetron Sputtered

$R_N^{mw}$, $\rho_N^{mw}$, $\delta_N^{mw}$ calculated at $T_c^{mw}$

@ column indicates temperature in K at which the $R_S^{eff}$ was measured.

thin films could be achieved by using "in-situ" deposition techniques. In our case, the fabrication of both the Bi-based and the Tl-based films included a post-deposition heat treatment. We have observed that $YBa_2Cu_3O_{7-\delta}$ thin films preparation processes which include a post-deposition annealing, such as sequential evaporation, yield films which are of lower quality than those deposited by "in-situ" techniques such as laser ablation.

The merits of our experimental technique have been tested against widely accepted characterization techniques, such as resonant cavity measurements. The consistency of the $R_s$ values measured using both techniques supports the validity of our method (see Fig. 7). The

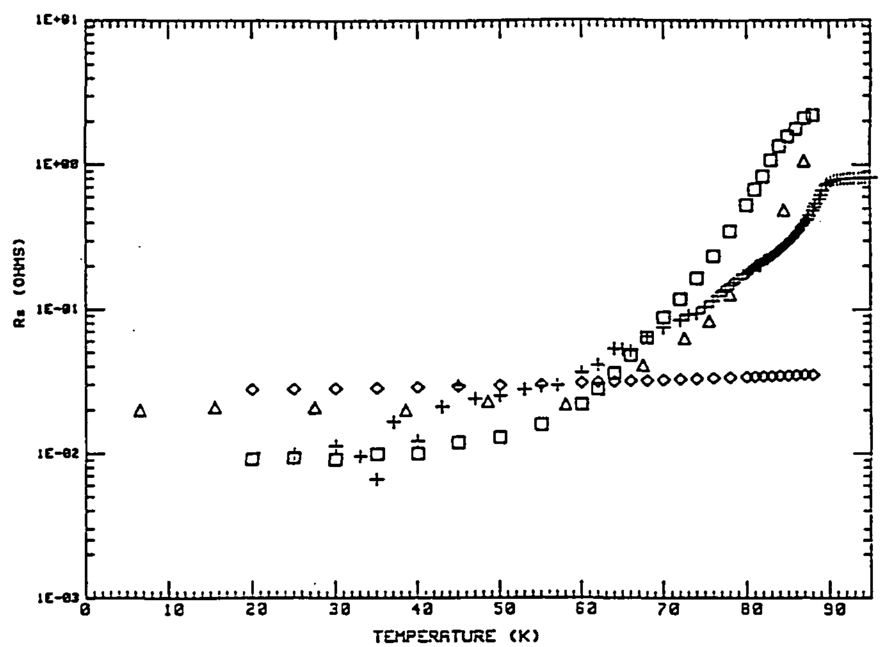


Figure 7: Surface resistance, $R_s$, versus temperature at 36 GHz for a $YBa_2Cu_3O_{7-\delta}$ thin film (4000 Å) on $LaAlO_3$ as measured by a microwave power transmission method (+) and by a cavity wall replacement method; Naval Research Laboratory (□) and Fort Monmouth (Δ). The $R_s$ for copper (◊) is also plotted for comparison.

strength of our technique is that it allows the calculation of several transport parameters, such as $\lambda$, $\sigma^*$, and $R_s$, from one single measurement, an attribute rarely found in any of the other probing techniques actually employed in HTS films research. The versatility of our technique rests not only in yielding values for $\lambda$, $\sigma^*$, and $R_s$ in good agreement with those obtained by other techniques, but also in being sensitive to the intrinsic anisotropy of these materials, as evidenced by the results for $\lambda$ of $YBa_2Cu_3O_{7-\delta}$ thin films.

# V. References


1. J. G. Bednorz and K. A. Muller, Z. Phys. B **64**, 189 (1986).
2. M. K. Wu, J. A. Ashburn, C. J. Torng, P. H. Hor, R. L. Meng, L. Gao, Z. J. Huang, Y. Q. Wang, and C. W. Chu, Phys. Rev. Lett. **58**, 908 (1987).
3. S. Sridhar and W. L. Kennedy, Rev. Sci. Instrum. **59**, 531 (1988).
4. J. S. Martens, J. B. Beyer, and D. S. Ginley, Appl. Phys. Lett. **52**, 1822 (1988).
5. W. Ho, P. J. Hood, W. F. Hall, P. Kobrin, A. B. Harker, and R. E. Dewames, Phys. Rev. B **38**, 7029 (1988).
6. R. E. Glover and M. Tinkham, Phys. Rev. **108**, 243 (1957).
7. S. L. Lehoczky and C. V. Briscoe, Phys. Rev. B **11**, 3938 (1971).
8. N. M. Rugheimer, A. Lehoczky, and C. V. Briscoe, Phys. Rev. **154**, 414 (1967).
9. Hewlett Packard, Product Note no. **8510-1**, 10 (1984).

# VI. PUBLICATIONS


1. Miranda, F. A.; Gordon, W. L; Heinen, V. O.; Ebihara, B. T.; and Bhasin, K. B.: *Measurements of Complex Permittivity of Microwave Substrates in the 20 to 300 K Temperature Range From 26.5 to 40.0 GHz,* Advances in Cryogenic Engineering **35,** Plenum Publishing Corporation, 1593-1599 (1990).

2. Miranda, F. A.; Gordon, W. L.; Bhasin, K. B.; Ebihara, B. T.; Heinen, V. O.; and Chorey, C. M.: *Complex Permittivity of Lanthanum Aluminate in the 20 to 300 K Temperature Range From 26.5 to 40.0 GHz,* Microwave Opt. Tech. Lett. **3,** 11-13 (1990).

3. Miranda, F. A.; Gordon, W. L.; Bhasin, K. B.; Heinen, V. O.; Warner, J. D.; and Valco, G. J.: *Millimeter Wave Transmission Studies of $YBa_2Cu_3O_{7-\delta}$ Thin Films in the 26.5 to 40.0 GHz Frequency Range,* Superconductivity and Applications, Plenum Press, 735-748 (1990).

4. Miranda, F. A.; Gordon, W. L.; Bhasin, K. B.; and Warner, J. D.: *Millimeter-wave Surface Resistance of Laser-ablated $YBa_2Cu_3O_{7-\delta}$ Superconducting Films,* Appl. Phys. Lett. **57,** 1058-1060 (1990).

5. Miranda, F. A.; Gordon, W. L.; Eck, T. G.; Bhasin, K. B.; Warner, J. D.; and Jenkins, K. A.: *Millimeter Wave Surface Resistance of $RBa_2Cu_3O_{7-\delta}$ (R=Y,Eu,Dy,Sm,Er) Superconductors,* NASA TM-102571 (1990).

6. Miranda, F. A.; Bhasin, K. B.; Heinen, V. O.; Kwor, R.; and Kalkur, T. S.:*Microwave Conductivity of Superconducting Bi-Sr-Ca-Cu-O Thin Films in the 26.5 to 40.0 GHz Frequency Range,* Physica C **168,** 91-98 (1990).

7. Warner, J. D.; Bhasin, K. B.; and Miranda, F. A.:*Dependence of the Critical Temperature of Laser-ablated $YBa_2Cu_3O_{7-\delta}$ Thin Films on $LaAlO_3$ Substrate Growth Techniques,* Supercond. Sci. Technol. 3, 437-439 (1990).

8. Bhasin, K. B.; Warner, J. D.; Chorey, C. M.; Ebihara, B. T.; Romanofsky, R. R.; Heinen, V. O.; Miranda, F. A.; and Gordon, W. L.: *Microwave Conductivity of Laser Ablated $YBa_2Cu_3O_{7-\delta}$ Superconducting Thin Films and Its Relation to Microstrip Transmission Line Performance,* NASA CP **3100,** 261-269 (1991).

9. Bhasin, K. B.; Warner, J. D.; Miranda, F. A.; Gordon, W. L.; and Newman, H. S.: *Determination of Surface Resistance and Magnetic Penetration Depth of Superconducting $YBa_2Cu_3O_{7-\delta}$ Thin Films by Microwave Power Transmission Measurements,* IEEE Trans. on Magn. **27,** 1284-1288 (1991).

10. Miranda, F. A.; Gordon, W. L.; Bhasin, K. B.; Heinen, V. O.; Warner, J. D.: *Microwave Properties of* $YBa_2Cu_3O_{7-\delta}$ *High Transition Temperature Superconducting Thin Films Measured by Power Transmission Methods,* Submitted for publication to Journal of Applied Physics, May 1991.