# Terahertz surface resistance of high temperature superconducting thin films

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We report on measurements of the surface resistance of  $YBa_2Cu_3O_x$  thin films at frequencies between 0.087 and 2 THz and temperatures between 50 and 120 K by time-domain terahertz-transmission spectroscopy (TDTTS) and resonant microwave spectroscopy. The determination of the surface resistance of superconducting thin films by TDTTS is extended to higher frequencies and thicker films than previously by numerically solving the complex transmission coefficient. The numerical solution also provides the dielectric function of the  $YBa_2Cu_3O_x$  thin films. The temperature and frequency dependence of the surface resistance of  $YBa_2Cu_3O_x$  thin films in the THz range is successfully explained by a weak coupling model of *d*-wave superconductivity which incorporates inelastic and elastic scattering. The surface resistance of  $YBa_2Cu_3O_x$  thin films at THz frequencies is compared to the surface resistance of gold and niobium. The advantages of  $YBa_2Cu_3O_x$  thin films for superconducting THz electronic devices are discussed. © 2000 American Institute of Physics. [S0021-8979(00)00806-9]

# I. INTRODUCTION

Since the discovery of high temperature superconductors (HTS) the application of these materials to high frequency electronics has received great interest.<sup>1</sup> Major advances in the implementation of HTS electronic devices at microwave frequencies<sup>2,3</sup> has tremendously increased the interest in the development of active and passive HTS electronic devices operating at terahertz (THz) frequencies. Promising applications of HTS THz electronics are, e.g., rapid-single-fluxquantum (RSFQ)<sup>4,5</sup> circuits, HTS transmission lines,<sup>6-9</sup> or antennas,<sup>10</sup> as well as Josephson junctions<sup>11</sup> as emitters and detectors of THz radiation. An important part of the design of these devices are numerical simulations<sup>12,13</sup> of their performance. The accuracy of the simulations relies on the input of precisely measured material properties of HTS at THz frequencies as well as on theoretical models describing the mechanism of HTS superconductivity. One of the key parameters describing the high frequency electromagnetic properties of HTS is the surface resistance because it determines the dissipation of ac currents. In this article an experimental and theoretical study of the THz surface resistance of  $YBa_2Cu_3O_x$  film is presented. The surface resistance data are compared to a theory of d-wave superconductivity. The advantages of  $YBa_2Cu_3O_x$  thin films for HTS THz electronic applications in comparison to normal conducting metals and conventional superconductors are discussed.

## **II. SAMPLES**

We have investigated an 80 nm thick c-axis oriented epitaxial  $YBa_2Cu_3O_x$  thin film which has been deposited by laser ablation<sup>14</sup> on (001) oriented MgO substrates  $(10 \times 10)$  $\times 1$ ) mm<sup>3</sup> in size. The superconducting transition temperature of the sample is  $T_c = 85.2$  K. The oxygen content of x  $= 6.95 \pm 0.05$  of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin film and the orientation of the *c*-axis of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>r</sub> with respect to the surface of the MgO substrate have been measured by Raman spectroscopy.<sup>15</sup> This analysis has demonstrated that  $\delta_c = 91\%$  of the volume of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> film exhibits *c*-axis orientation. The  $YBa_2Cu_3O_x$  film therefore exhibits good epitaxial quality. Moreover, the epitaxial quality of the sample is also demonstrated by the low residual surface resistance  $R_s = 1.84 \times 10^{-3} \Omega$  at T = 4.2 K and f = 0.087 THzas measured by resonant microwave spectroscopy<sup>16</sup> and compared to the results obtained by other researchers<sup>17</sup> which range from  $R_s = 0.3 \times 10^{-3} \Omega - 2 \times 10^{-3} \Omega$  for good quality films. Finally, the London penetration depth  $\lambda_T$ = 150.7 nm of our sample is close to the value  $\lambda_L \approx 140$  nm which is observed for high purity crystals.<sup>18</sup>

In order to get a picture of the kind of defects still present in our  $YBa_2Cu_3O_x$  thin film we have rescaled our

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surface resistance data taken at f=0.087 THz to f=0.010 THz using the relation  $R_s \propto f^2$  and compared it to the results of Ueno *et al.*<sup>19</sup> Recently, this group published a study which links defects in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin films, e.g., precipitates, stacking faults or grain boundaries with the surface resistance  $R_s$  at f=0.010 THz. The comparison of our data with the results of Ueno *et al.* reveals that the dominating defect in our sample are grain boundaries with grain size  $l_g=2 \mu m$ .

## **III. EXPERIMENTAL TECHNIQUES**

For the broadband measurement of the surface resistance of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin film two different experimental techniques were used. At f = 0.087 THz the surface resistance and the change in penetration depth  $\Delta\lambda(T)$  of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin film has been measured by resonant microwave spectroscopy. The experimental setup and the data analysis for this method have been explained in detail elsewhere.<sup>16</sup> At higher frequencies between 0.5 and 2.0 THz the surface resistance  $R_s(\omega, T)$  [Eq. (1)]<sup>20</sup> of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin film has been experimentally determined from measurements of the real and imaginary part of the dynamic conductivity  $\sigma(\omega,T) = \sigma_1(\omega,T) - i\sigma_2(\omega,T)$  as measured by timedomain THz-transmission spectroscopy (TDTTS),<sup>21–24</sup>

$$R_{s}(\omega,T) = \sqrt{\frac{\mu_{0}\omega}{2}} \left[ \frac{|\sigma|(\omega,T) - \sigma_{2}(\omega,T)|}{|\sigma(\omega,T)|^{2}} \right].$$
 (1)

During the last decade TDTTS has proved to be a unique experimental method for the broadband measurement of the surface resistance of HTS thin films at THz frequencies.<sup>21–24</sup> This technique measures the transmission of a picosecond electromagnetic transient through the superconducting film/ substrate combination  $E^F(t)$  and through a plain MgO substrate  $E^S(t)$  as a reference. The Fourier components of the transmitted  $E^F(\omega,T)$  and reference  $E^S(\omega,T)$ THz electric fields are obtained through fast Fourier transformation. They define the transmission as  $t(\omega,T) = E^F(\omega,T)/E^S(\omega,T)$ . As a phase-sensitive method TDTTS provides the complex transmission  $t(\omega,T)$ . In this case the complex index of refraction of the superconducting thin film  $n_2 = n + ik$  is related to the complex transmission  $t(\omega,T)$  through<sup>25</sup>

$$t(\omega,T) = \frac{E_1^F + iE_2^F}{E_1^S + iE_2^S} = \frac{4n_2}{(1+n_2)(n_2+n_3)\exp(-in_2\omega d/c) + (1-n_2)(n_2-n_3)\exp(in_2\omega d/c)},$$
(2)

where *d* is the thickness of the superconducting thin film, *c* is the velocity of light,  $n_3$  is the index of refraction of the MgO substrate, and  $\omega = 2\pi f$  the frequency.

Hitherto, the dynamic conductivity, from which the surface resistance  $R_s$  is calculated, has been extracted from the measured complex transmission [Eq. (2)] within the framework of the thin-film approximation<sup>21-24,26</sup> which assumes  $n_2\omega d/c \ll 1$  and  $\epsilon_1 \ll \epsilon_2$  so that  $n_2 \approx (\sigma/\omega \epsilon_0)^{1/2}$ . In our analysis the complex index of refraction of the superconducting thin film  $n_2 = n + ik$  is determined by numerically solving Eq. (2). Then the dielectric function  $\epsilon$  and the conductivity  $\sigma$  are calculated by dielectric conversion as  $n_2^2 = \epsilon = \epsilon_1 + i\epsilon_2$  and as  $\sigma = \sigma_1 + i\sigma_2 = i\omega\epsilon_0\epsilon$ . The real and imaginary part of the index of refraction and the dielectric function of

the  $YBa_2Cu_3O_x$  thin film as obtained by our measurements and analysis are displayed in Figs. 1 and 2.

In Fig. 3 the magnitude of the argument  $g = |n_2| \omega d/c$  of the exponential function which is expanded in the thin-film approximation as  $\exp(\pm in_2 \omega d/c) = 1 \pm in_2 \omega d/c$ , is calculated as a function of frequency for various film thicknesses *d* and  $|n_2| = 160$  which corresponds to a temperature of T = 53 K. The condition of the validity of the thin-film approximation is set as  $(n_2 \omega d/c)^2/2 \le 0.1$  or  $g \le 0.45$  and is indicated by a bold line in Fig. 3. The comparison of the numerical solution of Eq. (2) to the commonly employed thin-film approximation  $n_2 \omega d/c \le 1$  reveals that the determination of the THzconductivity of superconducting thin films by TDTTS is extended to thicker films and to higher THz frequencies.



FIG. 1. Experimentally determined real and imaginary part of the complex index of refraction  $n_2 = n + ik$  of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin film at THz frequencies.



FIG. 2. Real and imaginary part of the complex dielectric function  $\epsilon = \epsilon_1 + i\epsilon_2$  of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin film at THz frequencies obtained from the complex index of refraction through dielectric conversion  $n^2 = \epsilon$ .



FIG. 3. The modulus of the argument of the exponential function  $g = n_2 w d/c$  as a function of frequency for various thickness *d* of the superconducting thin films. The range of validity of the thin-film approximation is indicated by the bold line at g = 0.45.

Furthermore, by numerically solving Eq. (2) the entire complex dielectric function (Fig. 2) of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin film at THz frequencies is obtained. Hitherto, reliable measurements of the dielectric function of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> were only possible by far infrared (FIR) ellipsometry<sup>27</sup> at THz frequencies with a lower frequency limit of 3 THz for this method. Now, TDTTS expands the possibilities of measurements of the dielectric function of superconducting thin films below 3 THz.

#### IV. EXPERIMENTAL RESULTS AND DISCUSSION

The surface resistance of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin film as a function of temperature for various frequencies between 0.087 and 2 THz as measured by resonant microwave spectroscopy and TDTTS is displayed in Fig. 4. It is observed that the surface resistance exhibits a linear temperature dependence for temperatures well above the superconducting transition temperature  $T \gg T_C$ . In the vicinity above  $T_C$  the surface resistance  $R_s$  starts to deviate from this linear temperature dependence. For  $T < T_C$  the surface resistance  $R_s$  decreases strongly due to the transition in the superconducting state. This reduction of the surface resistance in the superconducting state is weaker for higher frequencies.

We describe the temperature dependence of the surface resistance of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin films at THz frequencies quantitatively by a weak coupling model of *d*-wave superconductivity. This theoretical description, which incorporates the *d*-wave symmetry of the order parameter  $\Phi = \Delta_0 \cos 2\phi$  has been extensively studied for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin films at f = 0.087 THz.<sup>16</sup> Our measurements of the surface resistance at frequencies above f = 0.087 THz are now testing the validity of this model at higher THz frequencies.

In our model the most important parameters describing the surface resistance of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin films at THz frequencies are the elastic scattering rate  $\Gamma_N^{\text{el}}$  and the inelastic scattering rate  $\Gamma^{\text{inel}}$ . It has been demonstrated that both inelastic and elastic scattering contribute to the total scattering within our range of temperatures. The elastic scattering rate  $\Gamma_N^{\text{el}}$  depends on the density of point defects  $n_{\text{imp}}$  which are the scattering centers and on the density of states N(0) at the Fermi level,



FIG. 4. Surface resistance of the  $YBa_2Cu_3O_x$  thin films at various frequencies between 0.087 and 2 THz. Experimental data are indicated by symbols, calculated theoretical curves are indicated by lines.

$$\Gamma_N^{\rm el} = \frac{n_{\rm imp}}{\pi N(0)} \sin^2 \delta_N.$$
(3)

Inelastic scattering is represented by a temperature dependent phenomenological scattering rate. Its temperature dependence is similar to the temperature dependence of inelastic scattering based on spin fluctuation exchange within the nested-Fermi-liquid<sup>28</sup> (NFL) model. While the NFL model takes the full frequency dependence into account, the phenomenological model neglects the frequency dependence involved in inelastic scattering,

.

$$\Gamma_{\rm phen}^{\rm inel}(T) = \Gamma^{\rm inel}(T_C) f_{\rm phen}(t), \qquad (4)$$

$$t = \frac{T}{T_C} \tag{5}$$

$$f_{\text{phen}}(t) = at^3 + (1-a)\exp\{b_1(t-1)[1+b_2(t-1)^2]\}.$$
(6)

The solid line in Fig. 4 is a fit of this theory to the measured surface resistance at f = 0.087 THz. The details of the calculations are explained in Ref. 16. The full set of fit parameters is summarized in Table I. Subsequently, the surface resistances of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin film at f = 0.5 THz, f = 1 THz, and f = 2 THz has been calculated with these fit parameters and are indicated in Fig. 4 as broken lines.

The only fit parameter which had to be adjusted separately for frequencies between 0.5 and 2 THz is a temperature independent constant  $R_0$  which is subtracted from the calculated surface resistance. The values of  $R_0$  are listed in Table II.  $R_0$  is explained by a temperature independent residual conductivity  $\sigma'_0$ . The residual conductivity  $\sigma'_0$  is attributed to extrinsic losses caused by grain boundaries present in our sample.

TABLE I. Summary of the fit parameter for the theoretical description of the surface resistance of the  $YBa_2Cu_3O_x$  thin film.

Elastic scattering rate $\Gamma_N^{\rm el}$	Scattering phase shift $\delta_N$	Inelastic scattering rate $\Gamma^{\text{inel}}(T_C)$	London penetration depth $\lambda_L$	$b_1$	$b_2$	а
1.2 meV	0.4 p	35.8 meV	150.7 nm	15	1	0.11

TABLE II. Temperature independent constant  $R_0$ .

$f (THz)  R_0 (\Omega)$	0.087	0.5	1.0	2.0
	0.32	0.25	0.3	1.0

Apart from this, the comparison of experiment and theory exhibits an excellent agreement of the temperature and frequency dependence of the surface resistance of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin film up to the highest measured frequency of f=2.0 THz. Thus, our model of the surface resistance of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin films has been successfully tested and proved to be valid up to 2 THz.

In the past, numerous measurements of the surface resistance of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin films and crystals have been performed at frequencies below 0.1 THz.<sup>29</sup> In those experiments the temperature and frequency dependence of the surface resistance has been successfully explained by models incorporating *d*-wave superconductivity. Only a few measurements of the surface resistance of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin films above 0.1 THz have been performed to date.<sup>9,21–24,30</sup> In this previous work, the surface resistance is either described by a general-ized two-fluid model,<sup>21–23,30</sup> by Bardeen–Cooper–Schrieffer (BCS) theory<sup>23,24</sup> or Mattis–Bardeen theory<sup>9</sup> which employ s-wave symmetry of the superconducting order parameter. These descriptions have been used because the *d*-wave nature of HTS was not yet known and theories of d-wave superconductivity were not available yet. The limitations of these models for the description of the surface resistance of  $YBa_2Cu_3O_x$  thin films have been extensively discussed by our collaboration previously.<sup>16,22,24</sup> As an example we point out that these theories are not able to describe the experimentally observed frequency dependence of  $R_s$  at THz frequencies. Therefore, by introducing our weak coupling model of d-wave superconductivity to the theoretical description of the surface resistance at THz frequencies and demonstrating good agreement with experiments up to f=2 THz we provide a powerful tool for the calculation of the surface resistance of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thin films as a function of temperature at THz frequencies.

With regard to the application of  $YBa_2Cu_3O_x$  thin films in HTS THz electronics we now compare our experimental and theoretical results of the surface resistance of  $YBa_2Cu_3O_x$  thin films to data of the surface resistance of gold (Au) and niobium (Nb). Au has widespread use in very large scale integration (VLSI) manufacturing, whereas Nb is used in superconducting THz electronic devices operating below 0.7 THz and temperatures below 9.2 K. The surface resistance of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub>, Au and Nb are displayed as a function of frequency for T = 4.2 K and T = 77 K, the boiling point of liquid helium (LHe) and liquid nitrogen  $(LN_2)$ , respectively, as well as some intermediate temperatures in Fig. 5. The surface resistance data for  $YBa_2Cu_3O_r$  at T = 77 K and T = 53 K originates from our experiments. Below T = 53 K the surface resistance of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub> has been calculated according to our theory. The surface resistance data for Nb is from Ref. 17 and the data for gold from Ref. 9. The surface resistance of Au at 4.2 K is not displayed in Fig. 5. Due to the anomalous skin effect the surface resis-



FIG. 5. Surface resistance of  $YBa_2Cu_3O_x$  thin films at THz frequencies compared to the surface resistance of gold and niobium for various temperatures.

tance of gold only changes by a factor of 2-3 between 4.2 and 77 K. Therefore, it is only slightly lower than the surface resistance at 77 K for our range of surface resistances.

The comparison reveals that for cooling with liquid nitrogen (T = 77 K) YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> has a lower surface resistance than Au for frequencies below 0.1 THz and for cooling with liquid helium (T = 4.2 K) for frequencies below 0.5 THz. The surface resistance of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> is always higher than the surface resistance of Nb at T = 4.2 K as long as Nb is superconducting ( $f \le 0.7$  THz). Compared to gold YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> is a suitable material for electronic devices only within a very narrow range of frequencies (0.1–0.2 THz) and temperatures (10–50 K).

### **V. CONCLUSIONS**

In conclusion, we have measured the surface resistance of a YBa<sub>2</sub>Cu<sub>3</sub>O<sub>r</sub> thin film at THz frequencies at temperatures between 50 K < T < 120 K. We successfully explain our data at THz frequencies by a theory of high-temperature superconductivity which incorporates the model of *d*-wave superconductivity. Based on this theory we have calculated the surface resistance of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>r</sub> thin films at temperatures below 50 K. With our experimental and theoretical study of the surface resistance of  $YBa_2Cu_3O_x$  thin films we provide reference data for device simulation. We have compared the surface resistance of  $YBa_2Cu_3O_x$  thin films at THz frequencies to the surface resistance of Au and Nb in the temperature range between 4.2 and 77 K. We demonstrate that  $YBa_2Cu_3O_r$  is a better material than Au for HTS electronic devices only in a narrow frequency range between 0.1 and 0.2 THz and for temperatures between 10 and 50 K because of its lower surface resistance. Time-domain THztransmission spectroscopy which is used to determine the surface resistance of superconducting thin films at THz frequencies has been improved by numerically solving the complex transmission. By this approach the application of TDTTS to the investigation of superconducting thin films keeps pace with the recent advances in the generation and detection of THz radiation by time-domain spectroscopy.<sup>26,31</sup>

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