Surface impedance of Tl-2212 thin films at THz-frequencies

Max Khazan, Ingrid Wilke, and Christopher Stevens

***Abstract*—We report on first measurements of the surface resistance and the London penetration depth of $Tl_2Ba_2CaCu_2O_8$ thin films in the frequency range from 200 GHz to 1.0 THz at temperatures from 25 to 268 K carried out by time-domain Terahertz transmission spectroscopy. We observe in the superconducting state an increase of the surface resistance with the square of the frequency up to 0.9THz at 77K and above. At 78K and 0.9 THz the surface resistance is 1.6 O. Below 77K the surface resistance increases with the cube of the frequency. Finally, the surface resistance of $Tl_2Ba_2CaCu_2O_8$ is compared to that of $YBa_2Cu_3O_{6.95}$ and gold.**

***Index Terms*—superconductivity, thallium compounds, surface resistance, Terahertz**

I. INTRODUCTION

The success of the application of high-T_C superconducting (HTS) thin films in microwave electronic devices [1]-[3] now stimulates research on high-T_C superconducting devices operating at Terahertz (THz) frequencies. The most promising materials considered for this purpose are $Tl_2Ba_2CaCu_2O_8$ (Tl-2212) and $YBa_2Cu_3O_{6.95}$ (YBCO).

At microwave frequencies HTS thin films are commonly characterized by the frequency and temperature dependent surface resistance and the temperature dependent London penetration depth. Hitherto, experimental studies of the surface resistance of Tl-2212 films have been restricted to frequencies below 0.1 THz [4]-[11] and above 1 THz [12]. To our best knowledge the electromagnetic properties of Tl-2212 films are unexplored between 0.1 and 1 THz. In order to close this gap we perform broadband measurements of the frequency dependence of the surface resistance of a Tl-2212 thin film between 0.2 and 1.0 THz in the superconducting as well as normal conducting state by the means of time-domain THz-transmission spectroscopy (TDTTS). These measurements also provide the temperature dependence of the London penetration depth.

II. EXPERIMENT

A. The Spectrometer

The time-domain THz transmission spectrometer is schematically depicted in Fig. 1. It is operated according to the pump-probe scheme which is based on the following principle: The optical pulsed laser beam is split in two parts. One beam, which is called pump and normally is of higher intensity, is used to excite an emitter of pulsed THz-radiationand the other beam (probe) gates a detector. The delay between pump and probe pulses is varied and by this the time profile of a THz-pulse is measured. In our spectrometer THz-pulses are released by a dc-biased large aperture photoconductor (LT-GaAs) emitter and detected with a photoconductive dipole antenna which allows us to cover the frequency range from 0.1 to 1.5 THz. A detailed description of various THz-spectroscopic techniques can be found elsewhere [13].

B. Data Processing and Analysis

In order to obtain the complex transmittance of a HTS thin film two time profiles are measured [14]: The THz-pulse transmitted through the HTS thin film / substrate and the reference THz-pulse which is either a freely propagating pulse or one passed through the bare substrate.

Time-domain THz-transmission measurements yield the complex transmittance of the HTS thin film / substrate as a whole. If a THz-pulse passed through a bare substrate is taken as a reference, the complex relative transmittance is expressed by the following equation:

$$T^*(w)=\frac{E^*(w)}{E_{ref}{}^*(w)}=\frac{2n_f(1+n_s)}{(1+n_f)(n_f+n_s)e^{-in_f wd/c}+(1-n_f)(n_f-n_s)e^{in_f wd/c}} \quad (1)$$

In (1) $E^*(?)$, $E_{ref}{}^*(?)$, are the Fourier transforms of the THz-pulse transmitted through the HTS thin film/substrate and of the reference pulse. The index of refraction of the HTS thin film and of the substrate are $n_f = n + i\,k$ and n_s,

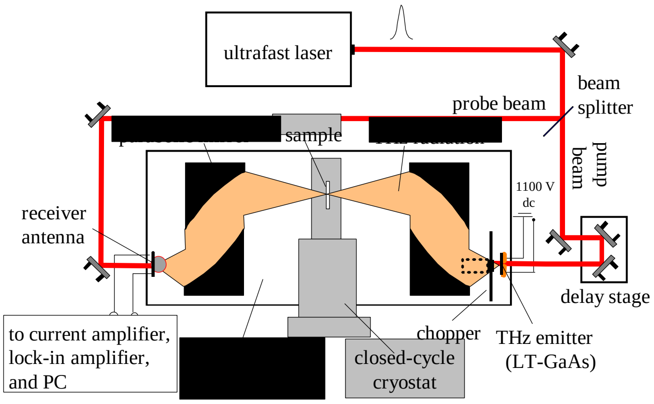

Fig. 1. Schematics of the experimental setup.

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M. Khazan and I. Wilke are with the Institut für Angewandte Physik, Universität Hamburg, Jungiusstr.11, D-20355 Hamburg, Germany.

C. Stevens is with the Department of Engineering Science , Oxford University, Parks Road, Oxford, OX1 3PJ, UK.

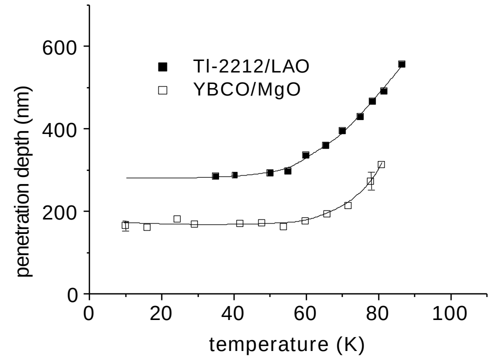

Fig. 2. London penetration depth of a $Tl_2Ba_2CaCu_2O_8$ (T_c=99K) and $YBa_2Cu_3O_{6.95}$ (T_c=85.2K) thin films.

respectively. The thickness of the HTS thin film is d and ? =2pf with frequency f. Equation (1) is solved numerically and n_f is obtained without making any *a priori* assumptions about the nature of the HTS thin film. The dielectric function e and the conductivity s of the HTS thin film are calculated as n_2^2=e'+ie'' and as i? e_0 e=s'-is'' .

Finally, the surface resistance is calculated as (2):

$$R_S(w,T)=\sqrt{\frac{m_0 w}{2}\frac{|s(w,T)|-s''(w,T)}{|s(w,T)|^2}} \qquad (2)$$

In order to determine the London penetration depths $?_L$ of the Tl-2212 thin films from conductivity spectra we have employed the generalized two-fluid model where the full conductivity s_{full} is presented as a sum of normal and superconducting contributions [2], [14]:

$$s_{full}(w)=s_n(w)+s_s(w)=\frac{n_n e^2 t/m^*}{1-iwt}+i\frac{1}{m_0 w\, l_L} \qquad (3)$$

The real part of the conductivity is then given by the expression:

$$s'(w)=\frac{s_0}{1+(wt)^2} \qquad (4)$$

where $s_0=n_n e^2 t/m^*$ is the dc conductivity and *f* is the ac electric field frequency. Thus, a parabolic fit to the $1/s'(f)$ spectra retrieves s_0 and the quasiparticle relaxation time t. Subsequently, the contribution of the quasiparticles to the imaginary part of the conductivity are determined by

$$s''_{Drude}(w)=\frac{s_0 w t}{1+(wt)^2} \qquad (5)$$

and then subtracted from the full s'' to obtain the contribution of the superconductive carriers:

$$s_{SC}(w)=s''_{full}(w)-s''_{Drude}(w)=\frac{1}{m_0\, l_L^2}\frac{1}{f} \qquad (6)$$

which is then fitted by a *1/f* hyperbola to extract the London penetration depth $?_L$.

C. *$Tl_2Ba_2CaCu_2O_8$ thin film*

The sample used for the measurements was a 80 nm $Tl_2Ba_2CaCu_2O_8$ film on a 10mm x 10mm x 0.5mm $LaAlO_3$ substrate. It has been deposited by sputtering a $BaCa_2Cu_2O_6$ precursor film in argon atmosphere and subsequently annealed at 877°C for 20 min in a sealed cru-cible containing optimal Tl-2212 powder. The c-axis of the $Tl_2Ba_2CaCu_2O_8$ is perpendicular to the surface of the substrate. Therefore, the conductivities in the ab-plane are measured in our experimental setup. The superconducting transition temperature of the $Tl_2Ba_2CaCu_2O_8$ film is T_C = 99 K. The temperature dependence of the penetration depth ?(T) as obtained from (6) is displayed in Fig.2. The London penetration depth extrapolated to zero temperature is $?_L(0)$=250 nm. We observe that ? ∝ T for T < 55K which indicates good crystalline quality of the $Tl_2Ba_2CaCu_2O_8$ film.The superconducting transition temperature as well as the London penetration depth of our sample are within the range of temperatures T_C=98 to 101K [4], [5], [9]-[12] and penetrations depths $?_L$=200 to 260nm [4]-[7], [11] which have been obtained by other researchers for Tl-2212 thin films deposited on $LaAlO_3$.

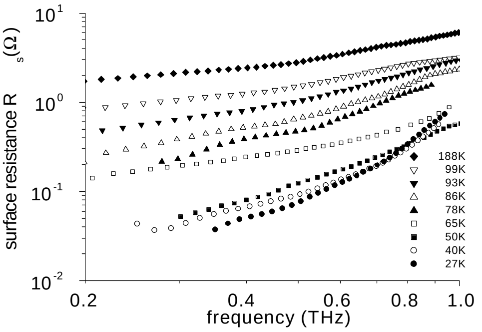

Fig. 3. Surface resistance of a $Tl_2Ba_2CaCu_2O_8$ thin film (T_c=99K) as a function of frequency for various temperatures.

III. RESULTS AND DISCUSSION

In this report we restrict our results to the frequency and temperature dependence of the surface resistance R_s(f,T). The full analysis of the experimental data within the framework of the generalized two fluid model will be published elsewhere [16].

A. *Frequency Dependence of the Surface Resistance*

The surface resistance from 0.2 to 1.0 THz of the Tl-2212 thin film is displayed in Fig.3 for temperatures above and below T_c.

We observe that the surface resistance exhibits a weakly nonlinear frequency dependence for T > T_c. For frequencies f below 0.5 THz the surface resistance is proportional to the square root of frequency as expected for the normal skin effect. For frequencies above 0.5 THz the surface resistance deviates from this behavior and linearly increases with frequency. As the Tl-2212 film enters the superconducting state the nonlinear frequency dependence of the surface resistance is strongly enhanced, particularly at higher THz-frequencies. In the superconducting state the frequencies below 0.8THz. Interestingly, above 0.8THz and below 65K

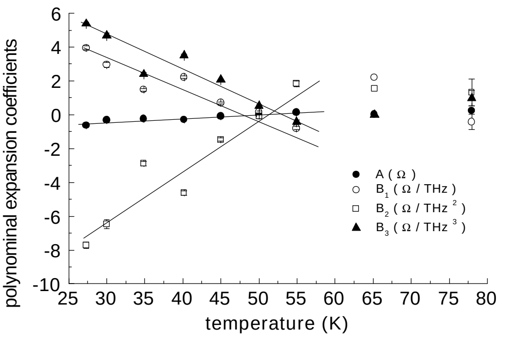

Fig. 4. Polynominal expansion coefficients A, B_1, B_2 and B_3 as a function of temperature for T =80K. Solid lines are guides to the eye.

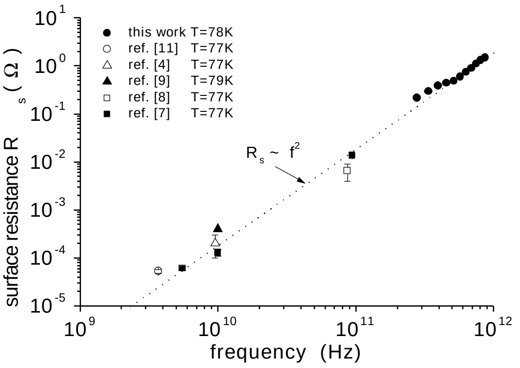

Fig. 5. THz- surface resistance at T=78K compared to previously published measurements of R_s of $Tl_2Ba_2CaCu_2O_8$ thin filmsperformed at microwave frequencies [4]-[11].

reversed such that it increases with lower temperatures.

In microwave studies of Tl-2212 thin films [7], [8] the frequency dependence of R_s has been described as increasing with the square of the frequency. We have found, however, that in the THz-range the surface resistance spectra cannot be satisfactorily fitted with a f^2 dependence. Thus, we fitted our data with a third-order polynom in frequency:

$$R_S(f,T)=A(T)+B_1(T)\,f+B_2(T)\,f^2+B_3\,(T)f^3 \qquad (7)$$

The temperature dependencies of the polynom coefficients A(T), B_1(T), B_2(T) and B_3(T) as determined by fits of (7) to the experimental data are displayed in Fig.4. We observe that A, B_1, B_2 and B_3 systematically change with temperature in the superconducting state. The frequency dependence of R_s is dominated by the linear term B_1 and the cubic term B_3 below T=60K. The coefficients B_1 and B_3 are highest for the lowest temperature measured and decrease with rising temperature whereas B_2 grows. At T=77K to T_c the frequency dependence of R_s is dominated by the quadratic term B_2. The new observation that R_s increases with lower temperatures for frequencies above 0.8THz and below 65K (Fig.2) is caused by the dominance of B_3 in this temperature range.

We now compare the THz-measurement of the surface resistance at 78K to previous measurements performed at frequencies between 3.7GHz and 94.1GHz and at temperatures T=77 to 79 K [4], [7]-[9], [11] (Fig.5). For this temperature range the largest amount of reference data is available. All previous measurements listed have been performed on Tl-2212 thin films deposited on $LaAlO_3$ substrates [4]-[10], [12] except reference [11] where MgO has been used as substrate. Although the experiments have been performed on TL-2212 films deposited by different methods, e.g. laser ablation [4], [5], [12] or sputtering [6]-[10], the various samples are rather uniform in their properties as characterized by transition temperatures in the range of T_C=98 to 110K and London penetration depths between $?_L(0)$=170 and 260nm.

In Fig. 5 it is clearly seen that our TDTTS measurements of R_s are in excellent agreement with previous microwave data with regard to the overall magnitude of R_s as well as the frequency depence $R_s \propto f^2$. Our broadband measurements confirm that the surface resistance R_s increases with the square of the frequency up to 1THz at T~77K. An increase of R_s with the square of the frequency is derived from (4) under the condition that s ''>> s ' and s '= s '(T) frequency independent which implies 2pf t <<1.

However, below T=77K R_s no longer increases with the square of the frequency but we observe $R_s \propto f^3$. This cannot be explained by s ' getting frequency dependent according to (4) because at higher frequencies it would make s ' to decrease and subsequently the rise of R_s to slow down (as illustrated for YBCO in Fig.7) or even to turn the surface resistance frequency independent. BSC analysis predicts a similar behavior of R_s or even its decrease at higher frequencies [17]. We point out that the deviation of R_s from the expected frequency dependence at higher THz-frequencies not only occurs in the superconducting but also in the normal conducting state. Therefore, we suggest that this behaviour is possibly caused by an additional loss mechanism, e.g. dielectric loss [18]. This hypothesis is currently under further investigation.

B. Temperature Dependence of the Surface Resistance

The temperature dependence of the surface resistance of the Tl-2212 thin film is shown in Fig.6. Above the superconducting transition ($T>T_C$) the surface resistance linearily depends on temperature for all frequencies. It monotonically drops two orders of magnitude between 99K and 27K with a slope of $R_s \propto T^4$ betwenn T = 70 K and 99 K and $R_s \propto T$ below T = 70 K at f=0.4THz. As frequency increases the surface resistance reaches a plateau at low temperatures (f=0.8THz) or even slightly increases (f=0.9THz). This is again the manifestation of the onset of $R_s \propto f^3$. The resulting residual surface resistance is 0.5O at f=0.9THz and T=30K.

C. Comparison of Tl-2212 with YBCO and Gold

At 77K gold always exhibits a lower surface resistance than Tl-2212 above 0.1THz (Fig.7). At the lowest temperature measured in our experiments (T=30K) Tl-2212 has lower

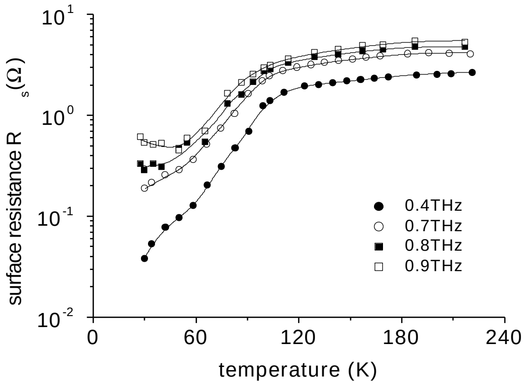

Fig. 6. Temperature dependence of the surface resistance of a $Tl_2Ba_2CaCu_2O_8$ thin film.

surface resistance than gold for f<0.5THz. Compared to YBCO, the R_s of Tl-2212 is of the same order of magnitude below 0.6THz at 77K. Above this frequency the surface resistance of Tl-2212 is higher than YBCO. So, it is seen that Tl-2212 films could be used in THz electronics with at least the same effectiveness as YBCO and thank to its higher T_c and lower 1/*f* noise [5] appears to be a very promising material for future applications.

IV. Conclusions

We have presented the first terahertz measurements of the surface resistance and the penetration depth of a Tl-2212 thin film. These measurements bridge the previously existing gap between 0.1THz to 1THz. We have carefully analysed the frequency dependence and observe that the surface resistance increases in the superconducting state with the square of the frequency at temperatures of 77K and higher i.e. exhibiting the same behavior as at microwave frequencies. At temperatures below 77K we have newly observed an increase of the surface resistance with the cube of the frequency.

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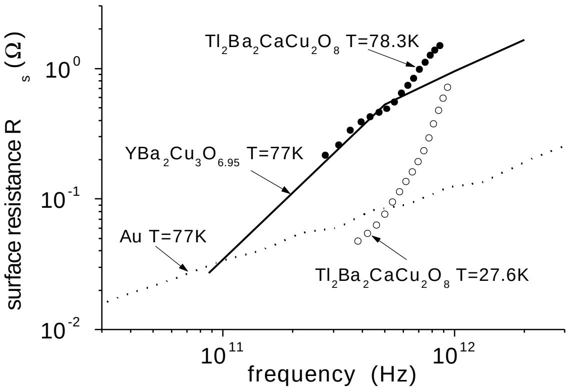

Fig. 7. Performance of $Tl_2Ba_2CaCu_2O_8$ compared to $YBa_2Cu_3O_{6.95}$ and gold. For $Tl_2Ba_2CaCu_2O_8$ R_s at 78.3 K is proportional to the square of the frequency up to 1THz. The data for $YBa_2Cu_3O_{6.95}$ are taken from [15].

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