# 80–111 GHz quasi-optical measurement of the complex conductivities of $YBa_2Cu_3O_7$ superconducting thin films

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(Received 29 June 1992; accepted for publication 11 December 1992)

Complex conductivities of $YBa_2Cu_3O_7$ high-temperature superconducting thin films were measured from 80 to 111 GHz using quasi-optical techniques. From transmission measurements, a peak was obtained in $\sigma_1$ versus temperature. Measurements of the frequency dependence of this peak position found it shifted to higher temperatures with increasing frequency. The data for both $\sigma_1$ and $\sigma_2$, as a function of frequency, were fitted using a BCS/effective medium model. This fit indicates that the observed peak in $\sigma_1$ is a result of the temperature dependence of the carrier mean free path combined with effects resulting from the granularity of the films.

The microwave and millimeter wave properties of high $T_c$ superconductors are of interest for practical device applications as well as for elucidating the fundamental nature of these materials. Measurements on the real part of the microwave conductivity, $\sigma_1$, as a function of temperature ($T$), carried out on various high $T_c$ materials, have revealed a narrow peak just below the transition temperature $T_c$.[1–9] This peak has been seen using various techniques and frequencies on thin films,[1–6,8] single crystals,[7,8] and powders.[9] To understand the origin of this phenomenon, it is necessary to make observations on a single sample over a range of millimeter wave frequencies. Here, we report on measurements that directly yield the real and imaginary parts of the conductivity, $\sigma_1$ and $\sigma_2$, of a thin film of $YBa_2Cu_3O_7$ (YBCO) from 80 to 111 GHz, and which demonstrate the frequency dependence of the $\sigma_1$ peak.

The values of $\sigma_1$ and $\sigma_2$ are obtained directly from the measured change in the amplitude and phase of a millimeter wave beam transmitted through a thin film. This method was the first to reveal the $\sigma_1$ peak, which was found at 60 GHz in Bi-Ca-Sr-Cu-O films[1] and in Tl-Ba-Ca-Cu-O films.[2] The experiments were repeated on YBCO films,[3] with the peak height and width varying from sample to sample. When the experiments were extended to higher quality films ($T_c \approx 88$ K), very narrow and reproducible peaks were measured.[4] In these and other studies, the peak was observed just below $T_c$ and was narrower for higher quality films. The peak has been seen mainly in the 30–60 GHz range,[1–4,6–9] with one report at 100 GHz[8] and one at 9 GHz.[5] A similar peak has been reported in a set of experiments at lower frequencies, 2–450 MHz[10] but at THz frequencies, the peak's existence is not clear.[11]

Several interpretations have been offered on the origin of the $\sigma_1$ peak.[1–16] To identify the nature of this peak and to place further constraints on possible interpretations, we carried out measurements to reveal the frequency dependence. Since the peak is known to vary from sample to sample, depending on film quality, we measured $\sigma_1$ at three frequencies using a *single* film. Because millimeter wave coherent leakage can affect such measurements (and is already the source of some possible confusion in the literature where phase shifts greater than 90° have been reported[5,6]), we focused on minimizing this leakage.

The YBCO superconducting thin films ($c$-axis oriented) with $T_c = 85$ K were synthesized using pulsed laser deposition.[4] A quasi-optical technique was used to measure the millimeter wave transmission loss and phase angle change of a film, versus both temperature and frequency. Transmission loss was measured by launching linearly polarized millimeter waves from a backward wave oscillator through a waveguide to a transmitting horn located in front of the samples. The power transmitted through the films was collected by a second horn located behind the sample and was recorded by a detector with a sensitivity of 0.10 nW. Four waveguide isolators were used to prevent interference between the source and the film, the film and the detector, and the source and the detector. Both the horns and the sample were installed in a vacuum chamber, and Brewster angle waveguide vacuum feedthroughs were used to seal the vacuum chamber. The temperature was stabilized to +/− 0.1 K. The source output frequency was measured using a source-locking frequency counter with kilohertz resolution. The change in phase of the transmitted wave through the sample was determined by balancing the signal in the sample and reference arms of a phase bridge with a resolution of +/− 1°.

The experimental success was due to the dynamic range of the measurement system and to the isolation from millimeter wave leakage across the measurement plane, which was achieved by sealing the perimeter of the samples with a thin layer of silver paint. An isolation of 70 dB was measured when the samples were replaced with a copper plate, also sealed using silver paint. Figure 1(a) shows the measured transmission loss and phase change across the $a$-$b$ planes of a 500 Å thick film on a 0.5 mm MgO sub-

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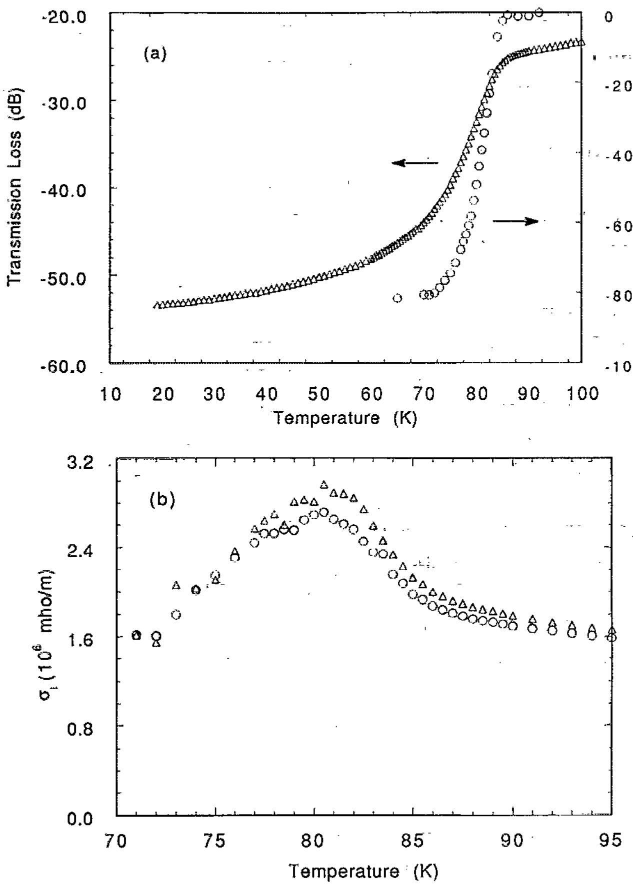


FIG. 1. (a) Transmission loss and phase change of a 500 Å thick $YBa_2Cu_3O_7$ film measured at 92 GHz; (b) $\sigma_1$ obtained from the data shown in (a) for a 500 Å thick film (circles), and also for a 750 Å thick film (triangles).

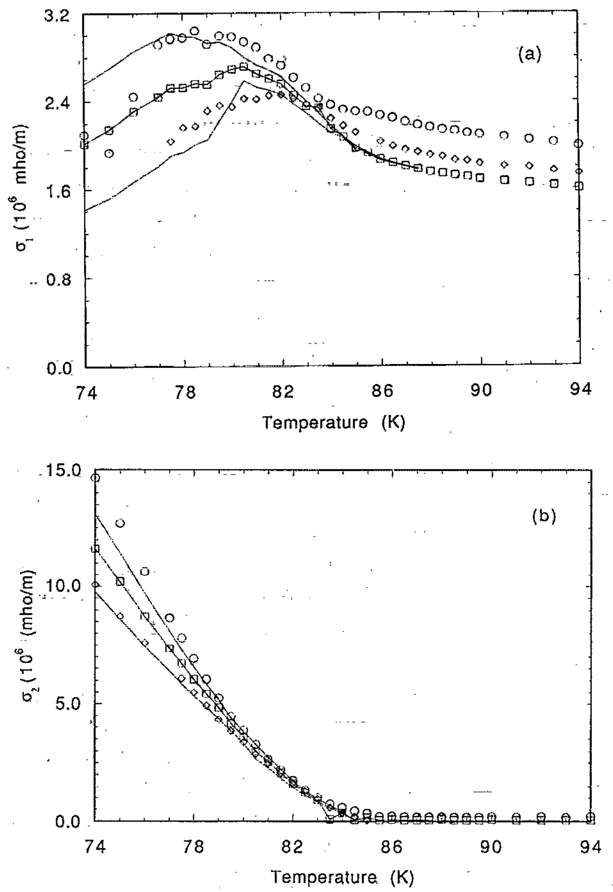


FIG. 2. Frequency dependence of (a) $\sigma_1$ and (b) $\sigma_2$. The points are from experimental data: ○=80.5 GHz, □=92 GHz, ◇=111.4 GHz, and the lines are calculated from the model.

strate with a 200 Å $SrTiO_3$ buffer layer, measured at 92 GHz. Above $T_c$, the transmission loss changes slowly following the trend of the dc resistivity. At $T_c$, the transmission loss drops rapidly followed by a slower roll off, resulting in a total change of −27 dB over the 90–19 K temperature range. There was no measurable phase change from 100 K down to $T_c$, and a phase change of 81.5° was measured from $T_c$ down to 72 K. Below 65 K, any additional change in phase was below our sensitivity levels.

The complex conductivities were calculated from the measured transmission loss and phase data. Figure 1(b) shows the real part of the conductivity calculated from the data in Fig. 1(a). As the temperature increased from 72 to 100 K, a peak was observed in $\sigma_1$ at 4.5 K below $T_c$. Also displayed in Fig. 1(b) is $\sigma_1$ for a 750 Å thick film at 92 GHz on a similar substrate. The two peaks for the two films have the same position and amplitude indicating that the peak in $\sigma_1$ was independent of film thickness. The calculated $\lambda$ and $R_s$ for the 500 Å film at 92 GHz was 1600 Å at 19 K and 10 mΩ at 72 K, respectively, which are consistent with published values for YBCO thin films.[4,17–19] The frequency dependence of the complex conductivity was investigated over the tuning range of the source. The measured $\sigma_1(T)$ and $\sigma_2(T)$ at 80.5, 92, and 111.4 GHz are shown in Figs. 2(a) and 2(b), respectively. Over the measurement range, the peak both narrowed and shifted to higher temperatures.

The data in Fig. 2 were analyzed with the BCS/effective medium model of Glass and Hall.[12] In this analysis, the expressions of Chang and Scalapino[20] for $\sigma_1$ and $\sigma_2$ (BCS theory with a finite, planar, mean free path, $l$) are equated to the experimental values, yielding two equations with two unknowns, namely $l/\pi\xi_0$ and $\sigma_n$. Here $\xi_0$ is the coherence length at $T=0$, and $\sigma_n$ is the normal state conductivity. Two free parameters in the theory are $\Delta_0/kT_c$ and $T_c$, where $\Delta_0$ is the gap energy at $T=0$. With the BCS value $\Delta_0/kT_c=1.76$, we found solutions using the 92 GHz data from the lowest $T$ point (74 K) up to some temperature $T_0<T_c$. Above $T_0$, where solutions no longer exist, the model assumes that we must describe a mixture of normal state and superconducting grains. In the present case, the value of $T_0$ was found to vary by 1 or 2 K depending on the choice of $T_c$ for 85 K$<T_c\leqslant$92 K. The results presented here correspond to $T_0=80$ K.

For $T>T_0$, the model invokes an effective medium theory (EMT) to describe the mixture of normal and superconducing grains with a new unknown, the fraction superconducting $f$.[12] An added parameter is the depolarization factor $g$. Following Ref. 12, we tried $g=0.15$ and $g=1/3$ (with no important differences in the results). Solutions for $f<1$ were found, from $T=T_c$ down to $T_0$.

Next we assumed that the results for $l(T)/\pi\xi_0$, $\sigma_n(T)$, and $f(T)$, which gave an exact fit to the 92 GHz data, are independent of frequency over the range of experimental frequencies (a reasonable assumption since $h\nu \ll \Delta$). We put these functions back in the model for $\sigma_1$ and $\sigma_2$ and evaluated the conductivity at the other experimental frequencies. The results are shown by the lines in Figs. 2(a) and 2(b). There is qualitative agreement with the experiment in the behavior of $\sigma_1$: on increasing the frequency, the peak shifts to higher temperature, becomes narrower and decreases in height. The agreement is even better for $\sigma_2$.

The same procedure for the data in Ref. 4 at 60 GHz shows the $\sigma_1$ peak's evolution with frequency even clearer (since the peak is narrower for a higher quality film): the peak moves to higher $T$, narrows, and shrinks in amplitude as frequency increases. This is the same trend that was measured for the peak at rf frequencies.[10,14] When we repeated the fitting procedure at 92 GHz using a larger than BCS gap, namely $\Delta_0/kT_c=4.0$, to calculate $\sigma_1$ and $\sigma_2$ at 80.5 and 111.4 GHz, we obtained no frequency dependence.

It has been suggested that the $\sigma_1$ peak may be accounted for by BCS theory with a fixed value of $l$ and $\Delta_0/kT_c>1.76$, thus arguing that the peak is the BCS coherence peak for a larger than BCS gap.[8,15] If $\sigma_2$ is fit in this way, however, then the calculated $\sigma_1$ is completely off from the experimental peak. Furthermore, if the measured $\sigma_1$ peak is fit by increasing $\Delta/kT_c$, then the predicted $\sigma_2(T)$ disagrees completely with experiment.[12] To fit $\sigma_1$ and $\sigma_2$ simultaneously with a BCS model, for a given $\Delta_0$, requires $l$ to vary with $T$ and necessitates a mixture model near $T_c$. Our analysis confirms that finding. Within the confines of weak-coupling BCS theory, it is the increase in $l$, in the $l \gg \xi_0$ regime, that causes the rapid decreases in $\sigma_1$ on the low $T$ side of the peak. On the high $T$ side of the peak, the rise of $\sigma_1$ as the temperature falls can be, in part, due to the intrinsic behavior of $\sigma_1$ when $l \approx \xi_0$ and, in part, due to a mixture effect (field expelled from the superconducting grains being concentrated in the normal grains). This explains why the peak is seen in single crystals and in high quality films, and widens in lower quality films, following the breadth of the falloff in dc resistance. In this model the peak is not due to the BCS coherence factors—the coherence effect vanishes as $l \gg \xi_0$.

Recent papers have used Eliashberg strong coupling theory (SCT) to study coherence peaks in the nuclear spin relaxation (NMR) rate,[21] and in $\sigma_1$.[22,23] The $\sigma_1$ peaks calculated in Refs. 23 and 24 do not really resemble the low frequency peaks measured near $T_c$. For relatively weak coupling or not-too-clean materials, the SCT peaks are too broad and too low in $T$. For stronger coupling or cleaner materials, these peaks do narrow and move to higher $T$, but they shrink in amplitude below that of the measured peaks. In the stronger coupling (or very clean) limit where the SCT coherence peak in $\sigma_1$ completely vanishes, the very rapid falloff in $\sigma_1$ with decreasing $T$ calculated just below $T_c$ resembles our BCS results for $l \gg \xi_0$. In this limit the measured $\sigma_1$ peak might be explained in SCT, as in the BCS theory here, by introducing a granular mixture in the 2–5 K range below $T_c$, where the peak occurs. In this same limit of SCT, the NMR coherence peak correctly vanishes.[21] Furthermore, there are no major conflicts in interpretation between the SCT and the BCS phenomenologies. In the SCT, the coherence peak vanishes because inelastic scattering from phonons leads to an intrinsic quasiparticle damping and the smearing out of the density of states within the gap (an effect which decreases as $T$ is lowered). In qualitative agreement with SCT, the BCS phenomenology of Ref. 24 finds that the NMR peak vanishes because pair-breaking scattering freezes out on lowering $T$; while here the $\sigma_1$ peak exists because of the decrease in the phenomological damping $1/l$ on lowering $T$.

In conclusion, we have experimentally established the existence of a frequency dependence of the $\sigma_1(T)$ peak from 80 to 111 GHz. Our analysis using a BCS/EMT model to fit the measured $\sigma_1(T)$ and $\sigma_2(T)$ simultaneously, exhibits the measured frequency dependence and leads to an explanation of the $\sigma_1$ peak's origin without the coherence effect. This explanation is consistent with the absence of the NMR peak and with strong coupling theory.


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