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Frequency-dependent relaxation rate in superconducting $YBa_2Cu_3O_{6+\delta}$

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The submillimeter-wave $3\ \mathrm{cm}^{-1} < \nu < 40\ \mathrm{cm}^{-1}$ complex conductivity of the reduced $YBa_2Cu_3O_{6+\delta}$ film ($T_C = 56.5$ K) was investigated for temperatures $4\ \mathrm{K} < T < 300$ K and compared to the properties of the same film in the optimally doped state. The frequency dependence of the effective quasiparticle scattering rate $1/\tau^*(\nu)$ was extracted from the spectra. $1/\tau^*$ is shown to be frequency independent at low frequencies and high temperatures. A gradual change to $1/\tau^* \propto \nu^{1.75 \pm 0.3}$ law is observed as temperature decreases. In order to explain the observed temperature dependence of the low frequency spectral weight above T_c, the quasiparticle effective mass is supposed to be temperature dependent for $T > T_c$.

I. INTRODUCTION

It is now well established that the complex conductivity of high-T_c superconductors has a highly unconventional character.[1,2] Despite many experimental efforts an unresolved question remains: is the frequency dependence of the conductivity due to a single mechanism or a sum of completely different processes such as the Drude peak and the midinfrared absorption?[3] Phenomenologically it has been proved to be useful to present the conductivity data on the basis of the extended Drude model with frequency-dependent effective mass m^* and the scattering rate $1/\tau^*$.[1,4] The analysis of the infrared conductivity has revealed a linear frequency dependence of the scattering rate with a temperature-dependent offset value.[1] Complementary, microwave techniques also allow the determination of the effective scattering rate at much lower frequencies,[5] and the corresponding values can be considered to constitute the low-frequency limit of the scattering rate. It is therefore reasonable to assume that $1/\tau^*$ becomes nearly constant below some characteristic transition frequency, as was recently observed[6] in the case of c-axis conductivity in $Y_{1-x}Ca_xBa_2Cu_3O_{6+\delta}$. Although this assumption agrees well with most experimental data, it is extremely difficult to observe this crossover frequency experimentally. This is mainly due to the fact that both components of the complex conductivity $\sigma^* = \sigma_1 + i\sigma_2$ have to be measured with high accuracy for frequencies below $50\ \mathrm{cm}^{-1}$; a range which is rather difficult to explore with conventional infrared techniques. As the temperature decreases below T_c, the accurate determination of the scattering rate becomes even more complicated because of the influence of the superconducting condensate that has to be subtracted from the conductivity prior to the calculation of the scattering rate. As a result, even less information exists concerning the low-frequency behavior of the scattering rate below T_c.

In view of the problems outlined above, the method of the submillimeter spectroscopy may be able to provide the necessary frequency-dependent information. Recently, using the submillimeter spectrometer, we were able to obtain the complex conductivity of the optimally doped $YBa_2Cu_3O_{6+\delta}$ film ($T_c = 89.5$ K) and to directly observe the quasiparticle relaxation below T_c.[7] In the present article we report on the properties of the same film in an oxygen-reduced state. Compared to the previous experiments, a higher transmission value of the reduced sample allowed us to estimate the frequency dependence of the effective scattering rate. In addition, a broader temperature interval, in which both $\sigma_1(\nu)$ and $\sigma_2(\nu)$ were available, made it possible to determine the spectral weight of the Drude peak and, thus, to observe the temperature dependence of the effective mass of the quasiparticles.

II. EXPERIMENT

The optimally doped 81 nm thick $YBa_2Cu_3O_{6+\delta}$ film on the $NdGaO_3$ substrate[7] was oxygen depleted by heating up in a controlled oxygen atmosphere, with subsequent quenching to room temperature. X-ray diffraction showed the c-axis orientation of the film. The mosaic spread of the c-axis oriented grains, i.e., the variation of the c axis with respect to the substrate axis, was 0.19°. Additional four circle x-ray diffraction revealed full in-plane alignment of the a/b-axes parallel to the substrate axes. Four-point resistivity measurement yielded an onset of the superconducting transition temperature, $T_c = 56.5$ K, with the 10%–90% transition width of 2.9 K. The changes of the lattice constant and critical temperature with oxygen depletion gave an estimate of the oxygen content of the sample.[8] For our sample a value of $\delta = 0.7 \pm 0.1$ has been determined.

The transmission experiments in the frequency range $3 \leqslant \nu \leqslant 40\ \mathrm{cm}^{-1}$ were carried out in a Mach-Zehnder interferometer arrangement,[9] which allows both the measurements of transmission, and phase shift of a film on a substrate. The properties of the blank substrate were determined in a separate experiment. Utilizing the Fresnel optical formulas, the absolute values of the complex conductivity $\sigma^* = \sigma_1 + i\sigma_2$ can be determined directly from the observed spectra without any approximations.

The frequency-dependent scattering rate can be calculated

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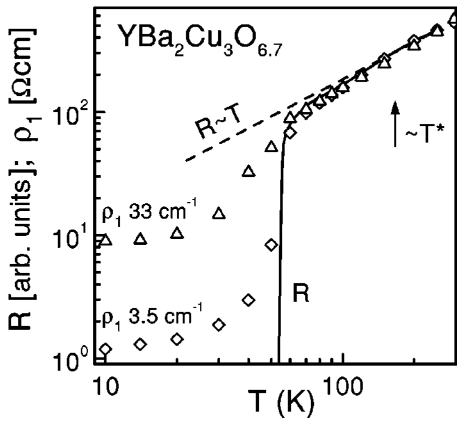

FIG. 1. dc resistivity of $YBa_2Cu_3O_{6.7}$ film compared to the data at submillimeter frequencies. Solid lines—four point dc resistivity, symbols—submillimeter-wave resistivity $\rho_1 = \sigma_1/(\sigma_1^2 + \sigma_2^2)$ at different frequencies. Dashed line indicates the linear temperature dependence of the resistivity.

from the complex conductivity using the modified Drude expression[1,4,10]

$$\sigma_D^* = \varepsilon_0 \omega_p^2 (1/\tau^* - i\omega)^{-1}, \tag{1}$$

where the plasma frequency ω_p and the renormalized scattering rate $1/\tau^*$ are assumed to be frequency dependent, $\omega = 2\pi\nu$ is the circular frequency, and ε_0 is the permittivity of free space. It should be noted that although the frequency dependences of ω_p and $1/\tau^*$ are a parametrization only,[11,12] the low-frequency limit of these functions can indeed be connected to the quasiparticle self-energy.[12–14]

III. RESULTS AND DISCUSSION

Figure 1 shows the four-point dc resistivity of the $YBa_2Cu_3O_{6.7}$ film as compared to the resistivity at submillimeter frequencies, $\rho_1 = \sigma_1/(\sigma_1^2 + \sigma_2^2)$. The dashed line represents the linear temperature dependence of resistivity. At temperatures $T < T^* = 200$ K the resistivity reveals significant deviations from a linear temperature dependence, which is a typical behavior for oxygen reduced $YBa_2Cu_3O_{6+\delta}$ known from literature.[15] The submillimeter and dc resistivity show the same temperature dependence within the experimental accuracy, which may be considered as an additional test of the film quality and homogeneity.

Figure 2 shows the frequency dependence of the complex conductivity of the $YBa_2Cu_3O_{6.7}$ film at different temperatures. The imaginary part of the conductivity is presented in form of a product $\sigma_2\nu$, which allows the determination of the superconducting spectral weight via[16,17]

$$\nu\sigma_2(\nu \to 0) = \frac{1}{2\pi}\frac{1}{\mu_0\lambda(0)^2} = \frac{1}{2\pi}\varepsilon_0\omega_{p,s}^2. \tag{2}$$

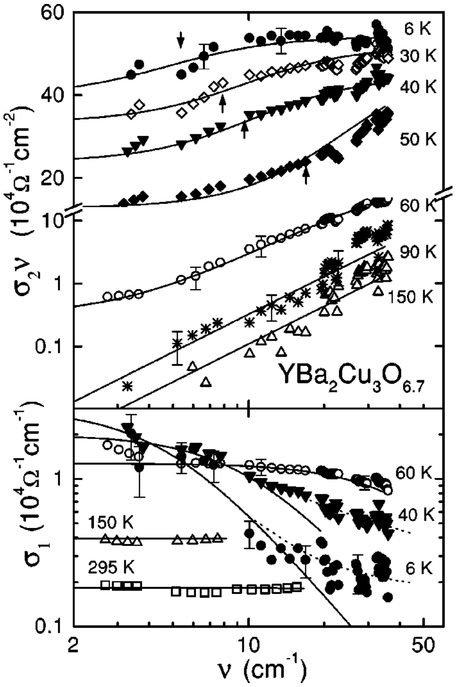

FIG. 2. Frequency dependence of the complex conductivity of $YBa_2Cu_3O_{6.7}$ film at different temperatures. Upper panel: the product $\sigma_2\nu$; lower panel: σ_1. Solid lines are fits according to Eq. (3). Dotted lines are drawn to guide the eye. Arrows indicate the approximate positions of the quasiparticle relaxation.

In Eq. (2) $\lambda(0)$ represents the low-frequency limit of the penetration depth, μ_0 is the permeability of free space, and $\omega_{p,s}^2$ is the spectral weight of the superconducting condensate. The real part of the conductivity (lower frame of Fig. 2) is frequency independent at high temperatures and increases with the decreasing temperature. At temperatures close to the superconducting phase transition a frequency dependence of $\sigma_1(\nu)$ is observed, which becomes significant as the temperature is lowered further. At high temperatures the imaginary part of the complex conductivity $\sigma_2\nu$ increases approximately as ν^2. As the temperature decreases below T_c, the low-frequency offset of $\sigma_2\nu$ becomes nonzero, which indicates the nonzero spectral weight of the superconducting condensate. The overall frequency dependence of $\sigma_2\nu$ shows then a minimum at zero frequency, with a characteristic width that becomes smaller with decreasing temperature. In analogy to the complex conductivity data of the unreduced $YBa_2Cu_3O_{6+\delta}$ sample[7] and according to theoretical predictions,[17] the width of this minimum qualitatively corresponds to the effective scattering rate. In order to obtain quantitative informations about the quasiparticle scattering and spectral weight, the conductivity was analyzed using the simple two-fluid model with frequency independent parameters.[18] However, as both σ_1 and σ_2 are measured as absolute values, we do not use the assumption $\omega_{p,n}^2(T) + \omega_{p,s}^2(T) = \omega_{p,s}^2(0)$. This assumption leads to $\omega_{p,n}^2(0) = 0$, i.e., all quasiparticles are condensed at zero temperature. As will be seen below, the normal spectral weight remains nonzero even at the lowest temperature of our experiment. The final expression for the complex conductivity at $\omega \neq 0$ can be written in the form

$$\sigma^*(\omega) = \sigma_D^* + \sigma_s = \varepsilon_0\omega_{p,n}^2(1/\tau^* - i\omega)^{-1} + i\varepsilon_0\omega_{p,s}^2/\omega, \tag{3}$$

where $\omega_{p,n}^2$, $\omega_{p,s}^2$ and $1/\tau^*$ are frequency *independent*, $\omega_{p,s}^2$ represents the spectral weight of the superconducting condensate (Eq. 2), $\omega_{p,n}^2$ is the spectral weight of the nonsuper-

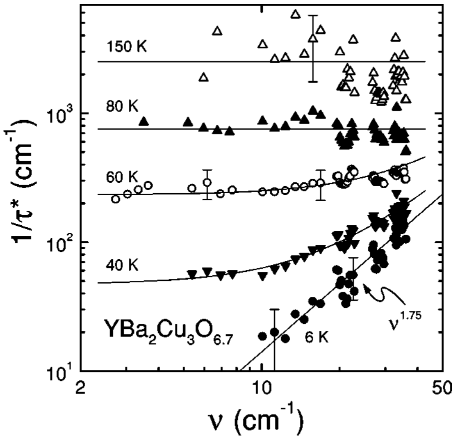

FIG. 3. Frequency dependence of the quasiparticle scattering rate of the reduced YBa$_2$Cu$_3$O$_{6+\delta}$ film at different temperatures. Solid lines are guides to the eye.

conducting component, and $1/\tau^*$ is the characteristic scattering rate. The results of the simultaneous fitting of the real and the imaginary parts of conductivity with $\omega_{p,n}^2$, $\omega_{p,s}^2$ and $1/\tau^*$ as free parameters are represented as solid lines in Fig. 2 and provide reasonable description of the experimental data. Prominent deviations are observed below T_c and at high frequencies. The experimental data have a significantly weaker frequency dependence compared to the Drude model. Therefore, a frequency-dependent scattering rate has to be used in order to fit the experimental data correctly. The deviations become less apparent in $\sigma_2\nu$ at low temperatures due to the dominance of the superconducting condensate [σ_s in Eq. (3)].

In order to obtain the frequency dependence of the scattering rate directly from the complex conductivity, we recalculated $1/\tau^*(\omega)$ from the experimental data using Eq. (1): $1/\tau^*=\omega\sigma_1(\omega)/\sigma_2(\omega)$. The procedure is quite straightforward for $T>T_c$ because $\omega_{p,s}=0$. For $T\leqslant T_c$ the term σ_s, describing the effect of superconducting condensate, has to be subtracted from the imaginary part of the conductivity. The obtained frequency dependence of the effective scattering rate is presented in the Fig. 3. The scattering rate is almost frequency independent at high temperatures in the submillimeter frequency range, as is well documented by the 80 K data set. A significant frequency dependence evolves below T_c with a crossover to a constant value. The crossover frequency shifts to lower frequencies as the temperature decreases. At $T=6$ K the scattering rate reveals a single power-law behavior and can be approximated by $1/\tau^* \propto \nu^{1.75\pm 0.3}$. This power law is close to the ν^2 behavior, which was observed in the normal state and at infrared frequencies in reduced YBa$_2$Cu$_3$O$_{6+\delta}$ sample;[19] however it is not clear, whether the same processes are determining the low-frequency electrodynamics above and below T_c.

TABLE I. Low-temperarure scattering rate of different samples of YBa$_2$Cu$_3$O$_{6+\delta}$ as obtained from submillimeter spectra (films) and microwave measurements (crystals).

No.	Sample	T_c(K)	$1/\tau^*$(cm^{-1})	Reference
1	crystal	93	4.7 ($\nu=0.14$ cm^{-1})	23
2	crystal	93	14 (1.2 cm^{-1})	23
3	crystal	88.7	1.7	24
4	film	89.5	45	7
5	film	56.5	43	this work

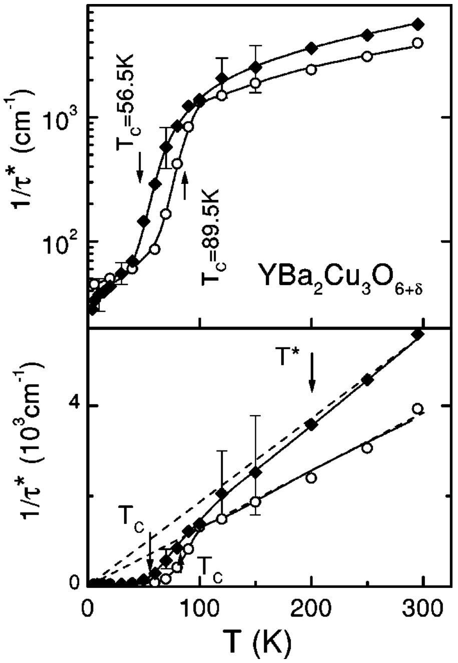

FIG. 4. Temperature dependence of the effective scattering rate of the reduced ($T_c=56.5$ K) and the optimally doped ($T_c=89.5$ K) (Ref. 7) YBa$_2$Cu$_3$O$_{6+\delta}$ film as extracted from the fits [Eq. (3)] to the complex conductivity data. Lower frame shows the data on the linear scale. Solid lines are guide to the eye. Dashed lines represent an extrapolation of the linear temperature dependence of the scattering rate observed at high temperatures.

Figure 4 shows the temperature dependence of the scattering rate of reduced ($T_c=56.5$ K) and optimally doped ($T_c=89.5$ K) films as determined using the simple Drude analysis [Eq. (3)] of the complex conductivity. These data may be considered as the averaged submillimeter-wave values of the scattering rate. The dotted lines in the lower frame indicate the possible linear temperature dependence of $1/\tau^*(T)$. Recently, Ioffe and Millis[20] proposed a model that implies a quadratic rather than linear temperature dependence of the scattering rate above T_c. Unfortunately, the present data do not allow a direct proof of this dependence, as the scattering rate above 150 K for both samples has been obtained assuming a temperature independent Drude spectral weight $\omega_{p,n}^2=\text{const}(T>150\ \text{K})$. As will be seen below, this assumption may have substantial influence on the form of the $1/\tau^*(T)$ curve. Below 200 K the experimental results for the underdoped sample start to deviate from the dotted line. This effect is not observed for the optimally doped sample and can be attributed to the opening of the spin gap in the reduced sample[21] below a characteristic temperature $T^*>T_c$.

Recently we were able to show that the scattering rate of different samples of YBCO remains nearly universal if scaled to some fixed temperature above T_c.[22] The sample independent value of $1/\tau^*$ was found to be ≈ 730 cm^{-1}. In this context it is interesting to compare the low-temperature values of the scattering rate of our films with the microwave data on single crystals.[23,24] This comparison is given in Table

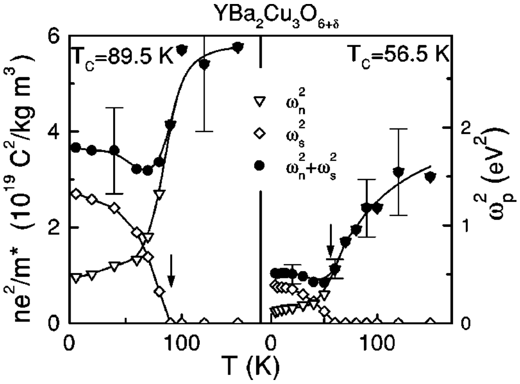

FIG. 5. Temperature dependence of the effective Drude spectral weight of the reduced ($T_c = 56.5$ K, right frame) and the optimally doped [$T_c = 89.5$ K, left frame (Ref. 7)] $YBa_2Cu_3O_{6+\delta}$ film. The data are extracted from the low frequency offset of $\sigma_2 \nu$ (superconducting component, $\omega_{p,s}^2$) and from the Drude fits [Eq. (3)] to the complex conductivity data (normal component, $\omega_{p,n}^2$).

I. The scattering rate for the thin films is approximately an order of magnitude higher than for the crystals, which probably indicates a higher level of defects in films. This conclusion correlates well with the strong sample dependence of the microwave scattering rate. However, comparing the crystal data at different frequencies (rows 1 and 2) we can also suggest an additional contribution from the frequency dependence of the scattering rate as discussed above (Fig. 3).

Figure 5 shows the temperature dependence of the Drude spectral weight for the optimally doped (left frame) and reduced (right frame) samples. The relatively high error bars reflect the uncertainties due to experimental errors and due to Drude-fits quality. The absolute value of the total spectral weight for the oxygen-reduced sample is lower compared to the optimally doped sample for all temperatures. This agrees well with the doping dependence of the total spectral weight as obtained by infrared measurements in the normal state[25] and by the magnetic penetration depth measurements in the superconducting state.[26,27] Figure 5 shows a strong temperature dependence of the spectral weight of a reduced sample above T_c. A possible explanation of this temperature dependence can be given in terms of a temperature-dependent effective mass of the quasiparticles, because the Drude spectral weight may be written as $\varepsilon_0 \omega_{p,n}^2 = ne^2/m^*$. According to the kinetic inductance data of Fiory *et al.*,[28] the quasiparticle concentration remains temperature independent above T_c. Therefore, our data suggest an increase of the effective mass of quasiparticles at low frequencies, approximately a factor of three, as the temperature is lowered from 150 to 60 K. These results may be compared with the two-component analysis of the infrared conductivity[1] carried out for frequencies above 50 cm^{-1}. This analysis revealed a nearly temperature independent weight of the Drude component for optimally doped $YBa_2Cu_3O_{6+\delta}$.[1,29] On the other hand, the analysis of the infrared conductivity on the basis of the modified Drude model (i.e., one-component analysis) reveals a remarkable temperature dependence of the effective mass in the low-frequency limit[1,4,14] both for underdoped and for optimally doped cuprates. Since both types of analysis are expected to coincide in the low-frequency limit,[4] the two results apparently contradict each other. Interestingly, our submillimeter data of the spectral weight are obtained by the simple Drude analysis of the first type, but provide the temperature-dependent effective mass similar to the low-frequency limit of the modified Drude analysis.

As the temperature is lowered through T_c, the normal spectral weight ($\omega_{p,n}^2$, Fig. 5) for both films decreases and then saturates at a finite value even at $T < 0.1\ T_c$. This decrease is followed by a gradual increase of the spectral weight of the superconducting component. As a result the full spectral weight for both samples reveals almost no changes for $T < T_c$ compared to $\omega_{p,s}^2 + \omega_{p,n}^2$ at $T = T_c$. This indicates that the apparent temperature dependence of the effective mass is ''frozen'' below T_c. Therefore, the two-fluid model assumption, $n_n + n_s = \text{const}$, which supposes a temperature-independent effective mass, holds for $T \leqslant T_c$. For higher temperatures, the conservation of the low-frequency spectral weight is violated due to the temperature dependent m^*.

IV. CONCLUSIONS

In summary, we have investigated the submillimeter-wave complex conductivity of a reduced $YBa_2Cu_3O_{6+\delta}$ film ($T_c = 56.5$ K) and compared it to the properties of the same film in the optimally doped state. Higher transparency and lower transition temperature of the reduced film allowed the observation of qualitatively new effects. The frequency dependence of the effective quasiparticle scattering rate has been extracted from the conductivity spectra. It was possible to show experimentally that the scattering rate is frequency independent at low frequencies and high temperatures. For decreasing temperature a transition between $1/\tau^* = \text{const}$ and $1/\tau^* \propto \nu^{1.75 \pm 0.3}$ is observed. In addition, the low-frequency spectral weight of the Drude component was estimated as a function of temperature and is shown to be temperature dependent above T_c. In order to explain the observed behavior, one has to assume an increase of the effective quasiparticle mass by a factor of three as the temperature is lowered from 150 K to 60 K. On the contrary, the total low-frequency spectral weight, $\omega_{p,n}^2 + \omega_{p,s}^2$, is temperature independent for $T \leqslant T_c$.

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