Electrodynamic properties of a superconducting film of Y–Ba–Cu–O in the submillimeter band

A. A. Volkov, B. P. Gorshunov, G. V. Kozlov, S. I. Krasnosvobodtsev, E. V. Pechen', O. I. Sirotinskiĭ, and Ya. Pettselt[1)]

Institute of General Physics, Academy of Sciences, USSR
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Direct measurements of the dynamic conductivity $\sigma_1(\nu)$ and dielectric constant $\varepsilon'(\nu)$ were carried out on a superconducting film of $YBa_2Cu_3O_{7-x}$ at frequencies from 8 to 32 cm^{-1} lower than the energy of the superconducting gap. It was established that in the normal phase $\varepsilon'(\nu,T)$ and $\sigma_1(\nu,T)$ show metallic behavior with a characteristic plasma frequency $\sim 3200\ cm^{-1}$ and a damping constant about 380 cm^{-1}. In the superconducting state, measurements of $\varepsilon'(\nu,T)$ agree with the classical premises of the low frequency electrodynamics of superconductors. The polycrystalline structure of the film exerts a greater distorting influence on the behavior of the dynamical conductivity. The temperature dependences of the concentration of the superconducting electrons, the penetration depth of the radiation, and the real and imaginary parts of the surface impedance are calculated.

INTRODUCTION

During the relatively short time since the discovery of high-temperature superconductivity (HTSC),[1,2] such a large number of works on the optical properties of the new materials have been published that it already exceeds the number of such works on the "classical" superconductors, beginning more than thirty years ago. Such a great activity in this field is related to the fact that modern optical research methods can give information about the properties of the electron and phonon subsystems, which is basic to the understanding of the mechanism of HTSC. There is particular interest in spectra in the range of photon energies of the order of the superconducting gap 2Δ and lower (frequencies $\nu < 2\Delta/h$), as it is just here that qualitative changes must occur in the electrodynamic characteristics of superconductors (conductivity σ_1 and dielectric constant ε'), brought about by the formation of the Bose condensate and by the discovery of the superconducting gap in the spectrum of the electronic states.[3–5] In reflection spectra, which are the basic object of measurement in infrared (IR) spectroscopy, these changes are manifested by increases of R at frequencies $\nu < 2\Delta/h$. However, since already in the normal state the reflection coefficient of a conductor is close to 100%, the problem of determining anomalies in the spectrum of the electronic states from reflection spectra is very complicated.

EXPERIMENTAL PART

In the present work, the basis of the experimental technique is the measurement of the transmission coefficient and of phase φ of a superconducting film on a dielectric substrate. This method has a higher sensitivity than the reflection method, but chiefly it permits the determination of ε' and σ_1 at each working frequency.

The complex transmission coefficient of this substrate-plus-film system is given by the expression[6]

$$t^* = \frac{T_{12}T_{23}T_{31}\exp\{i(\delta_2+\delta_3)\}}{1+R_{12}R_{23}e^{2i\delta_2}+R_{12}R_{31}\exp\{2i(\delta_2+\delta_3)\}+R_{23}R_{31}e^{2i\delta_2}}, \tag{1}$$

where the subscripts 1, 2, 3 refer respectively to the air, substrate and film; T_{pq} and R_{pq} $(p,q = 1,2,3)$ are the Fresnel transmission and reflection coefficients at the interface between the two media and depend on their refractive indices n and extinction coefficients k;

$$\delta_p = 2\pi\nu d_p(n_p+ik_p)/c \quad (p=2,\ 3)$$

d_p is the thickness of the substrate and film; $c = 3\times 10^{10}$ cm/sec. The values of t and φ are determined from experiment:

$$t=|t^*|^2, \tag{2}$$

$$\varphi=\arg(t^*). \tag{3}$$

Knowing (from previous measurement) the coefficients n and k of the substrate, it is possible, by solving the nonlinear equations (2) and (3), to determine the unknown coefficients n and k for the film, and from them to calculate the values $\varepsilon' = n^2 - k^2$ and $\sigma_1 = nk\nu$.

The measurement errors governed by the accuracy of the determination of t and φ, are, for $T = 300$ K and $T = 10$ K, respectively, ± 2000 and ± 6000 for ε' and ± 250 and ± 1000 $(\Omega\cdot cm)^{-1}$ for σ_1.

The region of applicability of formulas (1)–(3) is limited by the normal skin effect for conductors and by the local (London) limit for superconductors,[7–9] when the following relations hold for the penetration depth λ, the skin depth δ, the Ginzburg-Landau coherence length ξ, and the mean free path l: $l \ll \delta$, $\xi \ll \lambda$. According to Refs. 10–19, for $YBa_2Cu_3O_{7-x}$ single crystals, $\lambda_\parallel = 1800$ Å, $\lambda_\perp = 270$ Å, $\xi_\parallel = 4.3$–7 Å, and $\xi_\perp = 23$–31 Å (in directions parallel and perpendicular to the c-axis) and for ceramics $\lambda = 1000$–6200 Å, $\xi = 15$–22 Å, $l = 10$ Å. The quantity δ is obtained from the formula $\delta = (c/2\pi)\ (\sigma_1\nu)^{1/2}$: for $\sigma_1 = 10^3$ $\Omega^{-1}\cdot cm^{-1}$ and $\nu = 10\ cm^{-1}$ we get $\delta = 3\times 10^4$ Å. As we see, the conditions for locality are satisfied for superconductors with composition $YBa_2Cu_3O_{7-x}$.

As is well-known,[20–28] the most suitable substrates for growing high quality superconducting films of "123" composition are at present single crystals of $SrTiO_3$. However, this material is highly absorbing in the submillimeter (sbmm) band: $\varepsilon'' = 10$ ($T = 300$ K), $\varepsilon'' \gg 110$ ($T < 80$ K), due to an intense ferroelectric soft mode.[29] This circumstance complicates substantially the exploration of the properties of films on $SiTiO_3$ and limits their practical use.

As the substrate material we chose MgO which, accord-

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ing to our measurements, is characterized by low dielectric loss ($k < 10^{-4}$ at 300 K and 10 cm^{-1}) and by a practically constant refractive index over a wide temperature range (over the range from room temperature to helium temperatures it changed by about 1%). As regards the superconducting properties of films produced on MgO, they were somewhat inferior in their parameters to films on $SrTiO_3$.[22,23,25,30]

Films of composition $YBa_2Cu_3O_{7-x}$ were prepared by the method of pulsed laser sputtering, described in Ref. 30. The thickness of the film was measured by optical interferometry and amounted to 1300 Å $\pm 20\%$. X-ray diffraction showed that the film was polycrystalline, single phase, and textured with preferred orientation of the c axis perpendicular to the film. The inset of Fig. 3 below shows the temperature dependence of the magnetic susceptibility, characterizing a transition which begins at about 78 K and is 10–15 K wide.

The transmission coefficient and phase shift were measured with the submillimeter spectrometer "Epsilon"[31] in the region $\nu = 8$–$32\ cm^{-1}$ and in the temperature range 5–280 K. The parameters n and k were determined by computer solution of Eqs. (2) and (3).

RESULTS AND DISCUSSION

In Fig. 1 are shown typical $t(\nu)$ and $\varphi(\nu)$ spectra for a film of $YBa_2Cu_3O_{7-x}$ on an MgO substrate at two temperatures, above and below the critical $T_c = 73$ K. The solid lines show the result of the theoretical data reduction. The oscillations in the $t(\nu)$ spectra and the steps in the $\varphi(\nu)$ spectra are due to interference of the working radiation inside the plane-parallel substrate. In Fig. 2 are shown the temperature dependences of the transmission and phase shift for two frequencies, 9 and 21 cm^{-1}. In the normal phase, the transmission coefficient smoothly decreases with decreasing temperature, while the phase shift increases. Next, in the neighborhood of the phase transition, anomalies are seen in the $t(T)$ and $\varphi(T)$ dependences, namely, abrupt decreases of t and φ due to the transition of the film into the superconducting state. At $\nu = 9\ cm^{-1}$ the transmission coefficient decreases by an order of magnitude, while the phase shift decreases by more than a radian. At the higher frequen-

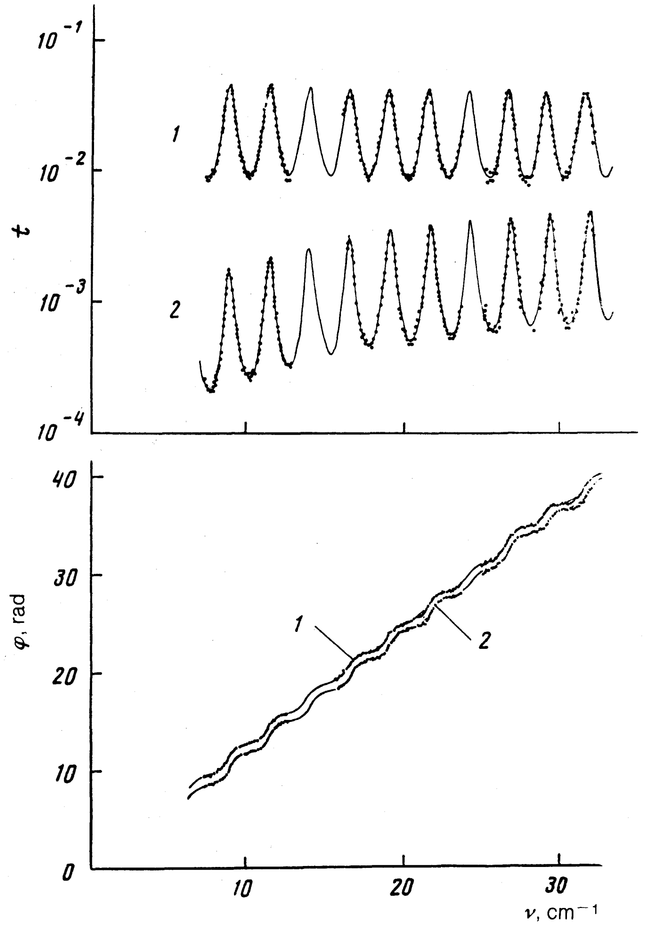

FIG. 1. Experimental spectra of the transmission coefficient and phase shift for a $YBa_2Cu_3O_{7-x}$ film on an MgO substrate measured at two temperatures: 1) 280 K $> T_c$, 2) 10 K $< T_c$. The solid curves show the results of a theoretical calculation by Eqs. (1)–(3).

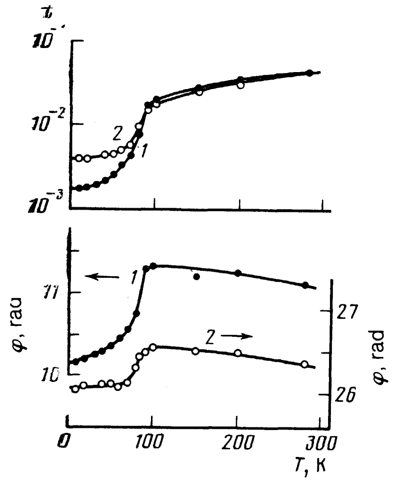

FIG. 2. Temperature dependences of the transmission coefficient and phase shift for a $YBa_2Cu_3O_{7-x}$ film on an MgO substrate, measured at two frequencies: 1) 9 cm^{-1}, 2) 21 cm^{-1}.

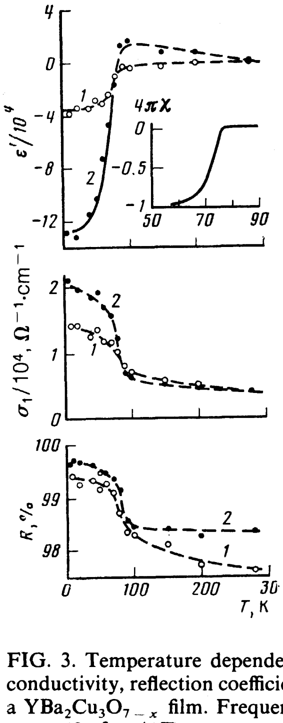

FIG. 3. Temperature dependences of the dielectric constant, dynamic conductivity, reflection coefficient, and magnetic susceptibility (inset) of a $YBa_2Cu_3O_{7-x}$ film. Frequencies: 1) 21 cm^{-1}, 2) 9 cm^{-1}. The solid curve (2) for $\varepsilon'(T)$ corresponds to theoretical calculations according to Eqs. (6)–(9), see the text.

cy (21 cm^{-1}), the absolute changes in t and φ at the phase transition are somewhat smaller, as shown in Fig. 2.

For these two frequencies, Fig. 3 shows the calculated temperature dependences of the parameters ε' and σ_1 and of the reflection coefficient

$$R=\frac{(n-1)^2+k^2}{(n+1)^2+k^2}\,. \qquad (4)$$

At the superconducting transition are seen a decrease of the dielectric constant and an increase of the conductivity and of the reflection coefficient. The greater changes again appear at the lower frequency 9 cm^{-1}. The quantity σ_1 increases by almost a factor of four, while ε' changes from zero at 280 K to enormous negative values of the order of 1.3×10^5 at helium temperatures. The reflection coefficient in the superconducting phase reaches 99.7%.

In Fig. 4 are shown the frequency dependences of the parameters of the $YBa_2Cu_3O_{7-x}$ film for $T=280\ \mathrm{K}>T_c$ and $T=10\ \mathrm{K}<T_c$. At high ($T>150$ K) temperatures there is practically no dispersion in the $\varepsilon'(\nu)$ and $\sigma_1(\nu)$ spectra ($\varepsilon'\approx 0$, $\sigma_1=(4\text{–}5)\cdot 10^3\ \Omega^{-1}\cdot\mathrm{cm}^{-1}$) and there is a decrease of reflection coefficient with increase of frequency. These are typical attributes of the Drude mechanism of metallic conductivity in the limit of low frequency $\nu\ll 1/\tau$, (where τ is the typical time between two successive collisions of carriers with phonons or impurities).[7] At $T<T_c$, dispersion appears in the $\varepsilon'(\nu)$ and $\sigma_1(\nu)$ spectra, particularly noticeable in $\varepsilon'(\nu)$.

According to classical ideas on the temperature and frequency dependence of the electrodynamic parameters of superconductors, the distinction between them and normal metals appears only for frequencies $\nu<2\Delta/h$.[5,32,33] Here the absorption of photons with energy $h\nu<2\Delta$, and therefore the magnitude of the dynamical conductivity σ_1 ($\nu\neq 0,T$), is determined by the concentration of unpaired electrons $N_n(T)$. Therefore at $T=0$, when $N_n=0$, the dynamic conductance vanishes for frequencies $0<\nu<2\Delta/h$. The condensate of Cooper pairs, which does not influence the dynamic conductivity for $0<\nu<2\Delta/h$, changes substantially the dielectric constant of the superconductor at these frequencies: with decreasing temperature, the negative contribution to ε' increases proportionally to the paired-electron concentration $N_1=N_0-N_n$ (N_0 is the total concentration). In the framework of the ideas of the two-fluid model of superconductivity and of local (London) electrodynamics, the temperature and frequency dependence of the complex conductivity $\sigma^*=\sigma_1+i\sigma_2$ of the superconductor can be presented in the form[8,9,32,33]:

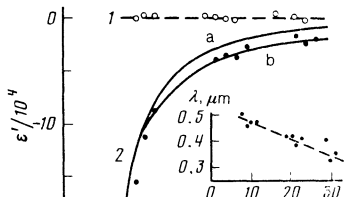

FIG. 4. Frequency dependences of the dielectric constant, dynamic conductivity, reflection coefficient, and London penetration depth (inset) of $YBa_2Cu_3O_{7-x}$ in the normal (1, $T=280\ \mathrm{K}>T_c$) and superconducting (2, $T=10\ \mathrm{K}<T_c$) states. The solid curves for $\varepsilon'(\nu)$ correspond to theoretical calculation by Eqs. (6)–(9) (see the text): a) $\lambda_0=0.5\ \mu$m, b) calculated for the frequency-dependent penetration depth (inset).

$$\sigma_1=N_ne^2\tau/m(1+\omega^2\tau^2), \qquad (5)$$

$$\sigma_2=\omega(\varepsilon_\infty-\varepsilon')/4\pi=c^2/4\pi\lambda^2\omega+N_ne^2(\omega\tau)^2/m\omega(1+\omega^2\tau^2) \qquad (6)$$

$$N_s/N_0=1-(T/T_c)^4, \qquad (7)$$

$$N_n=N_0-N_s=N_0(T/T_c)^4, \qquad (8)$$

where e and m are the charge and mass of the carriers, ε_∞ the contribution to ε' (or σ_2) from the higher frequency dispersion mechanisms $\omega=2\pi\nu$ is the cyclic frequency, and

$$\lambda(T)=(mc^2/4\pi N_se^2)^{1/2}=\lambda_0/[1-(T/T_c)^4]^{1/2} \qquad (9)$$

is the London penetration depth whose value λ_0 at $T=0$ is connected with the plasma frequency ω_p by the relation

$$\lambda_0^2=mc^2/4\pi N_0e^2=c^2/\omega_p^2. \qquad (10)$$

Turning now again to the experimental results (Figs. 3 and 4), we see that the behavior of $\sigma_1(\nu,T)$ does not fit the framework of classical ideas: with decreasing temperature and on transition to the superconducting phase, the absorption does not decrease but increases, and moreover there is a pronounced dispersion in the $\sigma_1(\nu)$ spectrum. This result (anomalous growth of the loss at $T<T_c$) agrees with the data of the microwave measurements on ceramic $YBa_2Cu_3O_{7-x}$ carried out at 9.4 GHz.[34] We assume that the presence of a low-frequency mechanism of absorption in this compound is due to weak intercrystallite coupling.

To test this hypothesis, we conducted by the same method a study of the submillimeter characteristics of weakly granular superconducting ($T_c=14$ K) films of NbN (thickness 1200 Å) on a fused-quartz substrate. We estimated the granularity, following Refs. 35 and 36, from the width of the resistivity $\rho(T)$ at $T\approx T_c$, from its semiconducting behavior at $T>T_c$, and from the absence of a characteristic maximum[5,35] at $T\approx T_c$ in the $t(T)$ dependence. The data obtained on $\varepsilon'(T)$ and $\sigma_1(T)$ of the NbN film are analogous in many ways to the results for the $YBa_2Cu_3O_{7-x}$ film, namely, a sharp decrease at $T<T_c$ was seen on the $\varepsilon'(T)$ dependence, but σ_1, on the other hand, did not tend to decrease on going to the superconducting state.

Let us turn now to the experimental results on the dielectric constant of the $YBa_2Cu_3O_{7-x}$ film in the superconducting state. Unlike the data on $\sigma_1(\nu,T)$, they agree well with the prediction of the classical theory: with decreasing temperature there is a decrease in ε' while the $\varepsilon'(\nu)$ frequency dependence is close to $1/\nu^2$ [see Eq. (6)]. Since at the same time the dielectric constant at low temperature ($T<T_c$) reaches anomalously large absolute values (of the order of 10^5 at $\nu=10\ \mathrm{cm}^{-1}$), one can conclude that its value at $T<T_c$ is determined mainly by the superconducting condensate, that is, it is described by the first term on the right-

hand side of (6). As is clear from Figs. 3 and 4, the $\varepsilon'(T)$ and $\varepsilon'(\nu)$ dependences (shown by solid lines) calculated by this equation with the use of (9) (for $\lambda_0 = 0.5$ μm, see below) actually describe the experimental data well. In the calculation of the $\varepsilon'(\nu)$ spectrum (Fig. 4, curve b), we took into account the dispersion of the London penetration depth λ. The $\lambda(\nu)$ dependence determined by Eq. (6) is shown in the inset of Fig. 4. With increasing frequency λ decreases, which also agrees with the theoretical predictions.[9,37,38]

Using (9) and (10), we calculated the temperature dependence of the penetration depth λ and of the concentration N_s of the superconducting electrons, as well as of the plasma frequency and of collision frequency[7]:

$$\gamma=1/\tau=\nu_p/2\sigma_1. \tag{11}$$

The $\lambda(T)$ dependence is shown in Fig. 5. It is clear that it can be described with the help of the empirical formula (9) with $\lambda_0 = 0.5$ μm. This value of λ_0 considerably exceeds the corresponding value for single crystals $\lambda_\perp = 270$ Å,[10] but is close to the data obtained for ceramic samples, $\lambda = 0.1$–0.38 μm.[13–19] In Fig. 5 is also shown the $N_s(T)$ dependence plotted on the assumption $N_0 = 10^{21}$ cm^{-3},[23,39,40] which corresponds according to (9) to an effective carrier mass $m = 9$ m_0,[17,24,30] where m_0 is the mass of the free electron.

For the plasma frequency and the collision frequency, we obtain ν_p(sbmm) $= 3200$ cm^{-1} and γ(sbmm) $= 380$ cm^{-1} for $\sigma_1 = 4\times10^3$ $\Omega^{-1}\cdot$cm^{-1}. The value of ν_p(sbmm) agrees with data of a number of papers on IR reflection from single-crystal and ceramic samples, $\nu_p = 3500$–4000 cm^{-1}.[39,41–43] According to Ref. 43 and our data $\gamma/\nu_p \ll 1$, that is, the carrier plasma is not overdamped.

On the basis of the obtained low-temperature data on ε'(sbmm), it is possible to evaluate the superconducting gap 2Δ with the help of the formula[33]

$$\sigma_2/\sigma_n=\pi\Delta \text{ th } (\Delta/2kT)/h\nu, \tag{12}$$

where σ_n is the dynamical conductivity in the normal (nonsuperconducting) state. For $\sigma_n = 6000$ $\Omega^{-1}\cdot$cm^{-1} we obtain $2\Delta = 40$ cm^{-1} ($T<T_c$) and $2\Delta(0)/T_c = 0.8$. Such a value for the energy gap seems unusually low. According to data on IR reflection, $2\Delta(0)/T_c = 1.6$–8.[21,24,39,41,44–52]

What experimental anomalies could be behind this difference? Aside from well known causes leading to a scatter in the experimental values of 2Δ, such as gap anisotropy[10,31,53] or distortion of the IR spectra by phonon and exciton modes, it is necessary in our case to point out one more—the polycrystalline structure of the film. As already remarked, it exerts such a strong influence on the conductivity in the superconducting phase that at frequencies below the gap ($h\nu<2\Delta$) an increase of σ_1 is observed instead of a decrease. Precisely for this reason, we based our calculations only on the $\varepsilon'(\nu_1 T)$ dependences, supposing that they are not distorted by the polycrystallinity. The grounds for such a conclusion were considered above. In our view, they are fairly persuasive but, of course, insufficient for categorical conclusions. At the same time one cannot fail to remark that our values of $2\Delta(0)/T_c$ are in surprising agreement with results obtained from IR absorption spectra.[54,55]

In conclusion, let us consider the characteristics of the investigated $YBa_2Cu_3O_{7-x}$ film, which may be of interest from the point of view of estimating the possibility of its use in the gigahertz band. In Fig. 6 are shown the temperature dependence of the real and imaginary parts of the surface impedance, $Z = Z_1 + iZ_2$, of the $YBa_2Cu_3O_{7-x}$ film, and also the skin depth δ, calculated from the formulas[5,32]

$$Z=(i8\pi^2\nu/c^2(\sigma_1+i\sigma_2))^{1/2}, \tag{13}$$

$$\delta=(c^2/4\pi^2\nu(\sigma_1+i\sigma_2))^{1/2}. \tag{14}$$

The observed variation of $\delta(T)$ in the neighborhood of T_c reflects a growth of the screening of the external field because of the supercurrent. On going to the superconducting phase, Z_1 is practically unchanged, in view of the increase in the active ohmic loss (Fig. 3). At the same time, the substan-

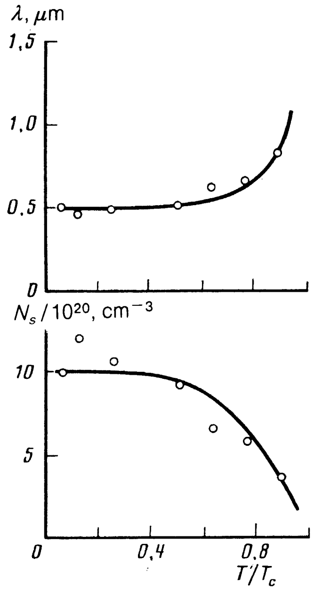

FIG. 5. Temperature dependences of the London penetration depth ($\nu = 9$ cm^{-1}) and of the concentration of superconducting electrons of the $YBa_2Cu_3O_{7-x}$ film. The solid lines show results of the calculation from Eqs. (9) and (10) at $\lambda_0 = 0.5$ μm.

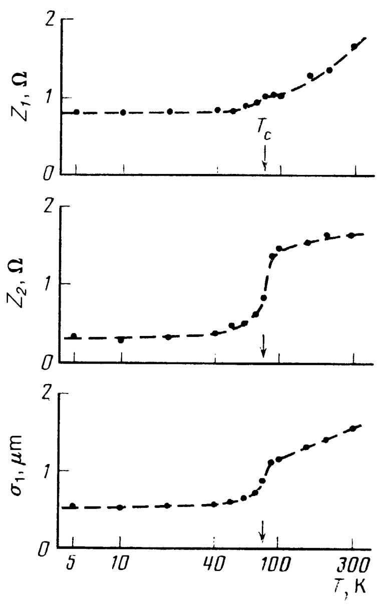

FIG. 6. Temperature dependences of the real (Z_1) and imaginary (Z_2) parts of the surface impedance and of the skin depth [Eqs. (13) and (14)] of $YBa_2Cu_3O_{7-x}$ film at $\nu = 9$ cm^{-1}.

tial change in the dielectric constant at $T<T_c$ leads to an appreciable change in the reactive part of the surface impedance Z_2. The absolute values of Z_1 and Z_2 of the investigated film are very large. An estimate shows that the surface impedance of copper at room temperature ($\sigma\approx 6\times 10^5$ $\Omega^{-1}\cdot\mathrm{cm}^{-1}$) is about 0.1 Ω, which is less than Z_1 and Z_2 of our film even in the superconducting state. The reason for this is the significant residual resistance resulting from the quasigranular structure of the superconducting $YBa_2Cu_3O_{7-x}$ film. It can be supposed that such a structure of the film is also the basic factor limiting the decrease of the Joule losses at $T<T_c$ in the microwave and millimeter bands in the case of the ceramic samples of high-temperature superconductors.[56–65]

CONCLUSIONS

The measurements of the submillimeter spectra of the dynamic conductivity σ_1 and dielectric constant ε' of a $YBa_2Cu_3O_{7-x}$ film on an MgO substrate allow the following conclusions.

1. In the normal ($T>T_c$) phase, the $\varepsilon'(\nu)$ and $\sigma_1(\nu)$ spectra have almost no dispersion, which is a characteristic attribute of metallic conductivity.

2. In the superconducting phase the dielectric constant decreases and can reach about -10^5, and the frequency dependence of $\varepsilon'(\nu)$ is close to $1/\nu^2$. These results agree with classical ideas about the electrodynamic properties of superconductors at frequencies $\nu<2\Delta/h$. At the same time, an anomalous increase is seen in the dynamic conductivity (absorption of radiation) at $T<T_c$ and $\nu<2\Delta/h$, which appears to be a consequence of the extra absorption of submillimeter radiation due to the polycrystalline structure of the film.

[1)]Institute of Physics, Czechoslovak Academy of Sciences.

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