Physica C 201 (1992) 13–20
North-Holland

PHYSICA C

Frequency dependent conductivity in $YBa_2(Cu_{1-x}Zn_x)_3O_7$ thin films

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Received 31 March 1992
Revised manuscript received 22 July 1992

The complex AC conductivity of $YBa_2(Cu_{1-x}Zn_x)_3O_7$ thin films has been investigated in the frequency range $10\,\mathrm{Hz} \leqslant \nu \leqslant 10^9\,\mathrm{Hz}$ and for temperatures $10\,\mathrm{K} \leqslant T \leqslant 300\,\mathrm{K}$. The influence of the substrate material on the complex conductivity is studied in detail. In the normal conducting state no intrinsic frequency dependence of the real part of the conductivity σ' could be detected, a finding which excludes a doping-induced localization of the charge carriers in this system. Below T_c, $\sigma'(T)$ reveals a coherence-like peak which has been predicted in the framework of the BCS theory. However, we propose that, in the doped superconductors, it is due to a distribution of T_c values. Finally, from the temperature dependence of the imaginary part of the conductivity we determine the T-dependence of the gap.

1. Introduction

Doping the superconducting cuprates with defects has proven to be a powerful tool to gain further insight into the unusual properties of these exotic materials. In particular, the substitution of the Cu-ions in $YBa_2Cu_3O_7$ by different transition metals (e.g. Co, Fe, Ni, Zn) has been studied in great detail [1]. Here we focus on Zn as dopant in the 1:2:3 superconductors. A preferential occupation of the Cu(2) plane sites by Zn has been reported by most authors [2]. However, this issue is still controversial. Doping with Zn leads to a substantial decrease of T_c (-10.5 K per 1% Zn [3]), and yields significant deviations from the linear "metal-like" temperature dependence of the resistivity ρ, commonly found for undoped samples. The mechanisms that lead to this behavior are still not completely understood. One possible explanation is that Zn doping promotes localization of the charge carriers [4,5]. Electrical transport in localized systems is due to hopping mechanisms [6]. Hopping conductivity is characterized by a power law behavior of the conductivity, namely $\sigma' \sim \nu^s$, with $s \leqslant 1$. Thus, measurements of the frequency dependence of the conductivity are an ideal tool to verify localization phenomena. Using this method, hopping conduction in high quality samples of $YBa_2Cu_3O_7$ has been excluded [7]. However, hopping is an important feature in the conductivity of the semiconducting "parent" compounds of high-T_c superconductors [8] and has also been observed in low quality samples of $YBa_2Cu_3O_{7-\delta}$ [9].

In addition, at temperatures $T \lesssim T_c$ measurements of the complex AC conductivity can give some clues about the mechanisms that lead to superconductivity in the cuprates. In BCS-type superconductors a so-called "coherence peak" is expected in the real part of the conductivity for frequencies well below the gap and for $T \leqslant T_c$ [10]. While some experiments on the complex conductivity below T_c have been conducted at microwave frequencies [11], data at audio and radio frequencies, especially of the frequency dependence of the peak, are still missing.

Here we report the frequency and temperature dependence of the complex conductivity $\sigma = \sigma' + i\sigma''$ [12] of $YBa_2(Cu_{1-x}Zn_x)_3O_7$ thin films. Above T_c, even at the highest Zn concentration examined, we find no evidence for a frequency dependence of σ' up to 1 GHz excluding hopping as a dominant mechanism for charge transport. Below T_c a peak in $\sigma'(T)$

is observed which, however, is not due to coherence effects.

2. Experimental details

Thin films of $YBa_2(Cu_{1-x}Zn_x)_3O_7$ with Zn concentrations $0.02 \leqslant x \leqslant 0.1$ were prepared by planar DC-sputtering under high oxygen pressure from single targets [3]. Various substrate materials have been used. To achieve good electrical contacts silver pads were evaporated onto the films and subsequently annealed in O_2 atmosphere [3]. Here we report results for $YBa_2(Cu_{1-x}Zn_x)_3O_7$ films with Zn concentrations $x=0.02$, 0.04 and 0.1 on $SrTiO_3$ substrates and with $x=0.04$ on MgO. The thickness of the films varied between 100 nm and 200 nm with an uncertainty of 50%. Measurements of the complex conductivity over eight decades of frequency were performed using two different experimental setups.

For frequencies $\nu \leqslant 10^7$ Hz an autobalance-bridge (HP 4192) was used which allows one to make measurements with a four-point contact geometry. The samples were mounted on the cold plate of a closed cycle refrigerator (CTI-Cryogenics) enabling measurements down to 10 K.

At higher frequencies (10^6 Hz $\leqslant \nu \leqslant 10^9$ Hz) data were recorded using an HP 4191A impedance analyzer connected to the refrigerator system via an air line [13]. In this type of reflectometric measurements the sample is mounted between two pins connected to the inner and outer conductor of the air line. An electrical connection between pins and the silver pads on the film is achieved by silver paint. Reflectometric methods require two-point contact configurations and the contact resistances contribute to the observed conductivities. In addition, significant contributions may arise from the inductance L of the silver paint and the pins, especially at the highest frequencies. This has to be accounted for by the use of suitable equivalent circuits in the evaluation of the data.

3. Results and discussion

Figure 1 shows the temperature dependent resistivity of three films on $SrTiO_3$ with various Zn concentrations. The decrease of T_c with increasing Zn doping levels is clearly demonstrated and is in accord with previous measurements [3,4]. At $x=0.04$ and, very clearly, at $x=0.1$, $\rho(T>T_c)$ deviates significantly from a linear temperature dependence with $\partial^2\rho/\partial T^2>0$. This could be interpreted as a localization effect. Only the sample with $x=0.02$ shows an almost linear behavior as it is commonly found for undoped $YBa_2Cu_3O_7$ samples. Remarkably, the three samples reveal very different slopes $d\rho/dT$. However, one has to keep in mind the uncertainties concerning the thickness of the films. Therefore the different slopes do not necessarily imply a violation of Matthiessens rule which has recently been reported to be valid in Zn doped $YBa_2Cu_3O_7$ superconductors [14].

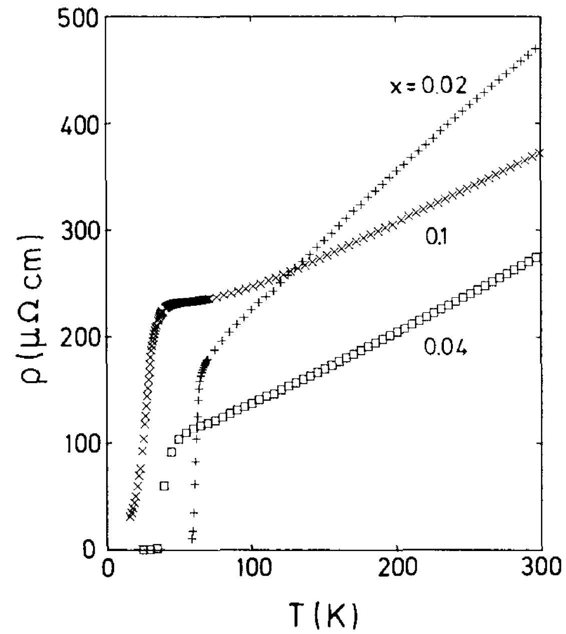

Fig. 1. Temperature dependence of the resistivity of $YBa_2(Cu_{1-x}Zn_x)_3O_7$ thin films on $SrTiO_3$ substrates for three different Zn concentrations: $x=0.02$ (+); 0.04 (□); and 0.1 (×).

The results on the films with $SrTiO_3$ as substrate material provide experimental evidence for a strong influence of the substrate on the complex conductivity. Hence, we also investigated $YBa_2(Cu_{1-x}Zn_x)_3O_7$ films on MgO substrates. The sample with $x=0.04$, which will be discussed later in detail showed a superconducting transition temperature $T_c=72$ K. The transition temperature is too high compared to values reported in literature [3,4]. Possible explanations for this behavior may be an incorrect Zn concentration and/or an inhomogeneous distribution of the dopant. The conductivity results on this sample clearly demonstrate that the influence of MgO as substrate material is negligible even at the highest frequencies investigated.

The frequency dependence of σ' for $T > T_c$ is shown in fig. 2 for a film with $x=0.1$ deposited on $SrTiO_3$. At all temperatures, σ' is almost frequency independent up to a frequency of 10^7 Hz. The slight deviations at the lowest frequencies are due to systematic errors of the autobalance-bridge at the edges of the available frequency range. However, above 10^7 Hz a strong frequency dependence is observed. Similar results are shown in fig. 3 for the real part of the conductivity in $YBa_2(Cu_{1-x}Zn_x)_3O_7$ with 4% Zn on MgO. However, even at a first glance the $\sigma'(\nu)$ behaves rather different for $\nu > 10$ MHz.

To demonstrate the different frequency dependences at high frequencies more clearly, figs. 4 and 5 show the real (top: (a)) and the imaginary part (bottom: (b)) of the conductivities for $x=0.1$ on $SrTiO_3$ and for $x=0.04$ on MgO, respectively. For the $YBa_2(Cu_{0.9}Zn_{0.1})_3O_7$ film on $SrTiO_3$ σ' exhibits a peak at temperatures below 200 K (fig. 4(a)). Correspondingly, σ'' passes through zero, revealing the characteristic features of a resonance curve (b).

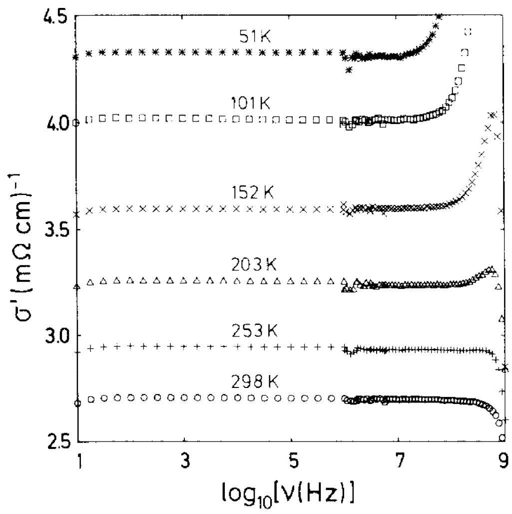

Fig. 2. Frequency dependence of the conductivity of a film with Zn concentration $x=0.1$ on a $SrTiO_3$ substrate at different temperatures (semilogarithmic plot): 298 K (○), 253 K (+), 203 K (△), 152 K (x), 101 K (□), 51 K (*).

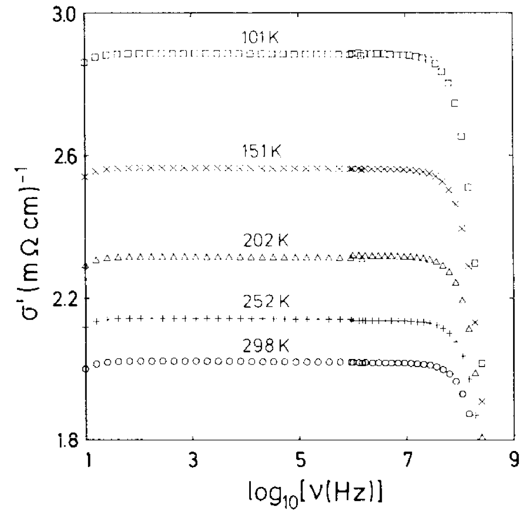

Fig. 3. Frequency dependence of the conductivity of a film with Zn concentration $x=0.04$ on a MgO substrate at different temperatures (semilogarithmic plot): 298 K (○), 252 K (+), 202 K (△), 151 K (×), 101 K (□).

These features are most pronounced at the lowest temperatures. We have observed a similar behavior for most films on $SrTiO_3$ substrates. On the contrary, the sample on MgO exhibits a different frequency dependence, showing, with increasing frequencies, a steplike decrease of σ' (fig. 5(a)) accompanied by a minimum in σ'' (b). The different behavior of $\sigma_{AC}(\nu)$ for $YBa_2(Cu_{1-x}Zn_x)_3O_7$ films on $SrTiO_3$ and MgO provides some experimental evidence that the measured high frequency conductivity is influenced by the substrate material. Indeed, $SrTiO_3$ has a strongly temperature dependent dielectric constant. The real part of the dielectric constant, ϵ', varies between 300 at room temperature and 20 000 at 4.2 K [15] while ϵ' of MgO is almost temperature independent and relatively small ($\epsilon' \approx 9.9$) [16].

Following these considerations we describe the data for films on $SrTiO_3$ substrates using the equivalent circuit indicated in fig. 4(a). Here the capacitance $C = \epsilon' \epsilon_0 C_0$ corresponds to the substrate being connected in parallel to the film. C_0 is the geometrical capacitance and ϵ_0 is the permittivity of the free space. It is sufficient to characterize the substrate by the real part of the dielectric constant, only. The loss ϵ'' of the substrate gives, in the most unfortunate case of high frequency and low temperature, rise to a resistance of some 1000 Ω [17] which can be ne-

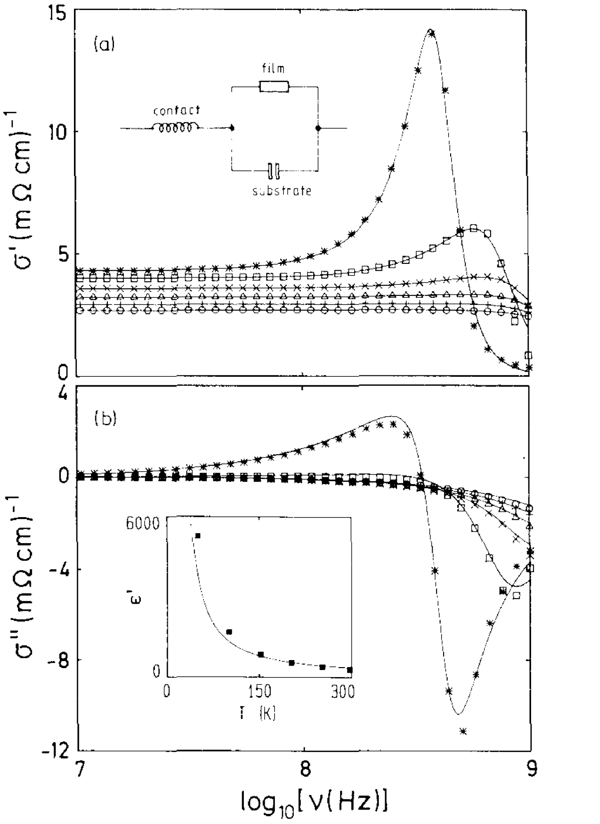

Fig. 4. Frequency dependence of the complex conductivity $\sigma=\sigma'+i\sigma''$ of a film with Zn concentration $x=0.1$ on a $SrTiO_3$ substrate for frequencies $\nu>10^7$ Hz. The real part σ' (a) and the imaginary part σ'' (b) are plotted vs. $\log_{10}\nu$: 298 K (○), 253 K (+), 203 K (△), 152 K (×), 101 K (□), 51 K (∗). The solid lines are the results of fits using the equivalent circuit indicated. The inset in (b) gives the temperature dependence of the dielectric constant ϵ' of the substrate as calculated from the fit parameter C (■) and the ϵ' of $SrTiO_3$ calculated after ref. [15] (solid line).

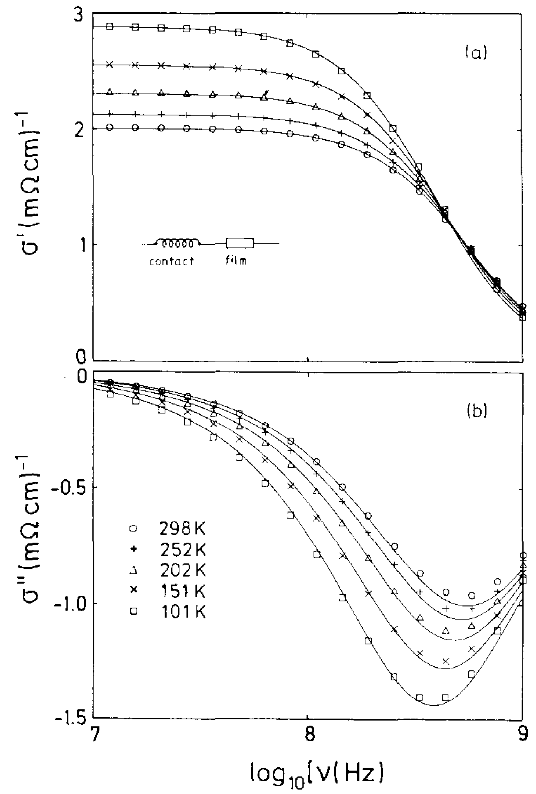

Fig. 5. Frequency dependence of the complex conductivity $\sigma=\sigma'+i\sigma''$ of a film with Zn concentration $x=0.04$ on a MgO substrate for frequencies $\nu>10^7$ Hz. The real part σ' (a) and the imaginary part σ'' (b) are plotted vs. $\log_{10}\nu$: 298 K (○), 252 K (+), 202 K (△), 151 K (×), 101 K (□). The solid lines are the results of fits using the equivalent circuit indicated.

glected being in parallel to a sample resistance of some 10 Ω. The inductance L not only accounts for inductive contributions of the film itself, but may also include the influence of the silver paint and contact pins as mentioned above. The lines in figs. 4(a) and (b) are the results of fits conducted simultaneously for the real and imaginary part using this equivalent circuit. During the fit procedure the values of the three elements have been treated as free parameters. The resulting temperature dependence of ϵ', is shown in the inset of fig. 4(b). The solid line in this figure has been calculated using the Barret formula $\epsilon'=M/[T_1/2)\ \coth(T_1/2T)-T_0]$ with $M=9\times10^4$ K, $T_0=38$ K and $T_1=84$ K which approximates the dielectric constant of $SrTiO_3$ [15]. The good correspondence of both curves strongly supports the validity of the equivalent circuit used. The inductance L was determined as $1.4(\pm0.1)$ nH, almost independent of temperature, and the resistance R follows the temperature dependence of the sample resistance as determined at low frequencies. We want to emphasize that it is possible to describe the data without assuming any intrinsic frequency dependent conductivity due to hopping processes.

$YBa_2(Cu_{1-x}Zn_x)_3O_7$ deposited on MgO can be described using an inductance in series with a purely ohmic resistance. This simple equivalent circuit (shown as inset in fig. 5(a)) provides a satisfactory fit to the data (solid lines in figs. 5(a) and (b)). L is temperature independent ($L=3.08(\pm0.02)$ nH) and R corresponds to the low frequency resistance of the sample.

Finally we want to give a short review of our results at temperatures below T_c. To exclude contributions of the substrate we restrict ourselves to films on MgO substrates. Figure 6 shows the temperature dependence of σ'((a), (b)) and σ''((c), (d)) for low and high frequencies. The real part of the conductivity exhibits a maximum below T_c and approaches a constant value at lower temperatures. The imaginary part σ'' drops abruptly at T_c and also approaches a constant (negative) value at low T. A maximum below T_c, the so-called coherence peak, has been predicted within the framework of the BCS theory to occur in σ' and also in the temperature dependence of the NMR relaxation rate $1/T_1$ [10]. It is a direct consequence of the correlations between scattering events due to the electron pairing and due to a strong increase of the density of states resulting from the formation of a gap. For "normal" superconductors this peak has commonly been found in NMR measurements (also called the Hebel–Slichter peak [18]). In $\sigma'(T)$ it has been observed only recently in superconducting Pb [19]. For high-T_c superconductors no coherence peak could be found in NMR measurements [20]. In contrast, a conductivity peak has been detected in some experiments at microwave frequencies which, however, revealed some deviations from BCS behavior [11]. The discrepancies between NMR and conductivity measurements have stimulated a number of theoretical investigations: Marsiglio [21] used that Eliashberg theory to calculate the dependence of the conductivity coherence peak on coupling strength, frequency and impurity content. However, within this BCS-like framework it was impossible to produce a coherence peak in the conductivity while producing none in the relaxation rate. In contrast, Nicole and Carbotte [22] found a peak in σ' using the marginal Fermi-liquid theory [23] also explaining the absence of a peak in

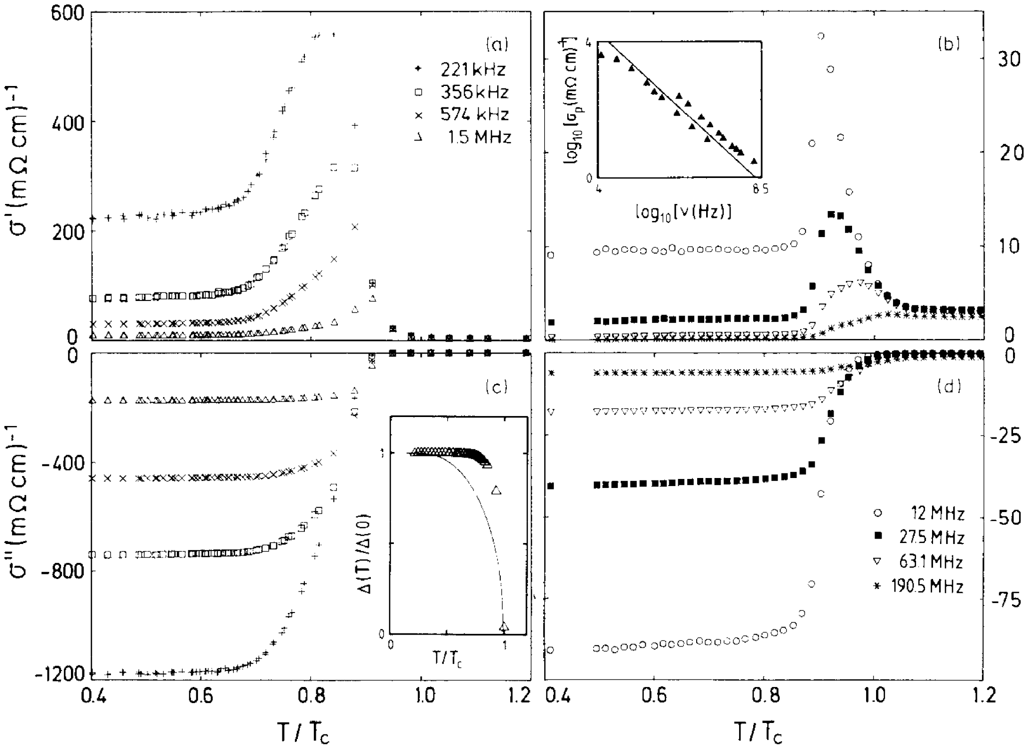

Fig. 6. Temperature dependence of the complex conductivity $\sigma=\sigma'+i\sigma''$ of a film with Zn concentration $x=0.04$ on a MgO substrate for low (a), (c) and high (b), (d) frequencies. The real part σ' (a), (b) and the imaginary part σ'' (c), (d) are plotted vs. the normalized temperature T/T_c: 221 kHz (+), 356 kHz (□), 574 kHz (x), 1.5 MHz (△), 12 MHz (○), 27.5 MHz (■), 63.1 MHz (▽), and 190.5 MHz (∗). The inset in (b) shows the frequency dependence of the peak height σ_p. The solid line indicates a slope of −1. The inset in (c) shows the temperature dependence of the superconducting gap energy, calculated from $\sigma''(T)$ (triangles). The solid line is the BCS prediction.

the NMR relaxation rate. Similar results have been obtained by Nuss et al. [24]. Recently it has been shown that fluctuation effects [25] or a distribution of T_c values [26] can also cause a peak in $\sigma'(T)$.

$\sigma'(T)$ in figs. 6(a) and (b) reveals a coherence-like peak which decreases with increasing frequencies and finally becomes suppressed for $\nu > 200$ MHz. The inset in fig. 6(b) shows the frequency dependence of the peak height σ_p. There is a slight offset between the low and high frequency measurement. However, for both measurements $\sigma_p(\nu)$ decreases like ν^{-1}, a behavior which has been predicted for thin films from fluctuation effects [27]. These predictions are valid for films with a thickness $d \ll \xi$ (ξ is the coherence length) which is not the case in our sample at the frequencies and temperatures under consideration. In bulk samples the frequency dependence of the peak maximum should follow $\sigma_p \sim \nu^{-0.5}$ [27]. In addition, fluctuations should lead to a peak exactly at T_c which is at odds with our findings. If the peak would be due to coherence effects, a logarithmic frequency dependence is expected [10], which again is in disagreement with the present data. Also the approach of a constant value of $\sigma'(T)$ at low temperatures (figs. 6(a), 6(b)) is at odds with BCS-theory [10].

Hence, we assume that a doping induced broadening of the superconducting transition is responsible for the occurrence of the peak in $\sigma'(T)$ [26]. In the normal conducting state and at not too high frequencies, the sample behaves like an ohmic resistor ($R \gg X = 2\pi\nu L$) and hence, the conductance $G' \approx 1/R$. At T_c the resistance decreases smoothly, while the reactance remains finite with values not too different from the normal conducting state (indeed, the superconducting phase transition is characterized by a phase jump of $-90°$ between current and driving field). Thus below T_c where $R \ll X$ the conductance behaves like $G' \approx R/X^2$. The maximum of the peak appears at the temperature where $R = 2\pi\nu L$. At very high frequencies X is of the order of R and the peak just below T_c becomes suppressed, a behavior that indeed is observed at GHz frequencies (see fig. 6(b)).

There are slight indications of fluctuation effects for $T > T_c$. This is supported by the observation of a much narrower peak exactly at T_c in undoped $YBa_2Cu_3O_7$ which shows a sharper transition [28]. This peak can satisfactorily be explained by fluctuation effects [28].

The temperature dependence of σ'' for various frequencies is shown in figs. 6(c) and (d). For $T \ll T_c$ the frequency dependence is well described by $\sigma'' \sim \nu^{-1}$, in accord with the BCS predictions. Within the BCS theory, for frequencies $h\nu \ll kT_c$, $\sigma''(T)$ is given by [10]:

$$\sigma''/\sigma_n = (\pi\Delta)/(h\nu)\tanh[\Delta/(2kT)] . \quad (1)$$

Here σ_n is the normal state conductivity and Δ the energy gap. Equation (1) allows one to perform the calculation of the temperature dependence of $\Delta(T)$ from the measured $\sigma''(T)$. A representative result (calculated from a four-point measurement of $\sigma''(T)$ at 1.5 MHz) is shown as inset in fig. 6(c) and compared to the BCS predictions [29]. This inset clearly reveals that close to T_c the temperature dependence of the gap is stronger than predicted by BCS theory and that saturation occurs already for $T/T_c < 0.8$. This behavior corresponds to the results from infrared and high-resolution electron-energy-loss studies [30] that the magnitude of the gap in $YBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_8$ does not appear to change with temperature almost up to T_c.

4. Conclusions

We have measured the complex conductivity of $YBa_2(Cu_{1-x}Zn_x)_3O_7$ thin films in the normal and superconducting state for Zn concentrations up to $x = 0.1$.

Above T_c no frequency dependence of σ' could be detected up to 1 GHz which excludes hopping conductivity as an important feature in the Zn doped $YBa_2Cu_3O_7$ superconductors. Therefore, theories assuming a localization of the charge carriers are not suited to explain the effects of Zn doping on the electronic transport properties of this system. In addition, the absence of hopping conductivity in all samples investigated seems to exclude the possibility that polarons or bipolarons play an important role in the formation of superconductivity [31].

Below T_c we found a peak in $\sigma'(T)$. The frequency dependence of the peak gives rise to serious doubts if it originates from coherence effects. Fluctuation effects alone also cannot account for its frequency and temperature dependence. We showed that the occurrence of this peak can easily be explained assuming a distribution of T_c values and our conclusion is, that coherence effects are absent in the electromagnetic absorption as well as in the NMR relaxation rate [20]. Finally, we extracted the temperature dependence of the energy gap from $\sigma''(T)$. We found a $\Delta(T)$ which develops sharply below T_c and is almost temperature independent for lower temperatures.

Acknowledgements

This research was supported by the "Bundesministerium für Forschung und Technologie" under contract number 13 N 5705/0 and by SFB252 (Darmstadt/Frankfurt/Mainz/Stuttgart). One of us (CT-R) was financially supported by the Conselho Nacional de Desenvolvimento Cientifico e Technologico do Brasil - CNPq.

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