Optical conductivity in YBa2Cu3O7−δ thin films

Andrei Pimenov, Alois Loidl, G. Jakob, H. Adrian

Angaben zur Veröffentlichung / Publication details:

Pimenov, Andrei, Alois Loidl, G. Jakob, and H. Adrian. 1999. "Optical conductivity in YBa2Cu3O7−δ thin films." *Physical Review B* 59 (6): 4390–93. https://doi.org/10.1103/PhysRevB.59.4390.

Optical conductivity in $YBa_2Cu_3O_{7-\delta}$ thin films

A. Pimenov and A. Loidl
Institut für Physik, Universität Augsburg, D-86135 Augsburg, Germany

G. Jakob and H. Adrian
Institut für Physik, Universität Mainz, D-55099 Mainz, Germany

(Received 16 October 1998)

The real and imaginary parts of the optical conductivity $\sigma_1 + i\sigma_2$ of partly untwinned $YBa_2Cu_3O_{7-\delta}$ thin films were measured by submillimeter spectroscopy for frequencies from 100 GHz to 1 THz. The frequency dependence of the conductivity below T_C can be described by a narrow Drude-like peak with strongly temperature-dependent relaxation rates. $\sigma_1(\nu)$ does not extrapolate to the universal *d*-wave value at 0 K. Impurity scattering significantly different to the unitary limit has to be assumed to achieve agreement with recent theoretical results. Including published infrared results we present $\sigma_1(\nu)$ in the spectral range from 0.1–100 THz. [S0163-1829(99)01605-7]

INTRODUCTION

The outstanding importance of measurements of the low-frequency electrodynamics ($\nu < 1$ THz) of high-temperature superconductors was unambiguously demonstrated during the last years.[1] Specifically, in addition to infrared (IR) (Refs. 2 and 3) experiments, microwave and terahertz-spectroscopy experiments were performed in the superconducting state. And while some experimental results provide experimental evidence of *d*-wave superconductivity, a number of open and so far unresolved questions remain: (i) the frequency dependence of the complex conductivity below 1 THz is not at all well characterized and it seems important to determine the spectral shape of the quasiparticle scattering well below T_C, (ii) the important role of impurities in *d*-wave superconductors is well known: not only the impurity concentration but also the scattering strength is of outstanding importance. Up to now for most of the samples investigated it is not known if unitary scattering or if the Born limit is the more appropriate approximation, and (iii) there seems to be no convincing experimental proof of the universal low-temperature dc conductivity, which has been predicted by P. A. Lee.[4]

A large body of work has been performed on $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin films and single crystals using microwave surface-resistance methods.[5–10] Important information also was gained employing terahertz spectroscopy[11–14] and by submillimeter measurements.[15,16] A detailed review on most of these results has been given by Bonn and Hardy.[1] The most probable resume of these experiments can be given as follows: (i) The peak below T_C which is observed in the real part of the optical conductivity σ_1, is due to two competing temperature dependencies. With decreasing temperature the normal-fluid density decreases, while the scattering time strongly increases.[6,11] (ii) At least in high-purity YBCO single crystals the penetration depth λ depends linearly on temperature. Theoretically it has been shown by Annett *et al.*[17] that a linear temperature dependence of the low-frequency penetration depth is a signature of a *d*-wave pairing state. Later on it has been shown by Hirschfeld and Goldenfeld[18] that a crossover temperature T^* exists below which $\lambda \propto T^2$. T^* crucially depends on the defect concentration, and this behavior indeed has been demonstrated experimentally by Bonn *et al.*,[8] and (iii) there is no real consensus concerning the superconducting gap Δ_0. But based on the low-temperature measurements of the penetration depth the estimates yield $\Delta_0 \approx 2-3 k_B T_C$.[1]

In this paper we report on measurements of the complex conductivity on high quality $YBa_2Cu_3O_{7-\delta}$ films in the frequency range from 100 GHz to 1 THz. The films were prepared by high-pressure dc sputtering from a stoichiometric $YBa_2Cu_3O_7$ target onto a (001) oriented $NdGaO_3$ substrate. A deposition time of 40 min resulted in an 81-nm-thick epitaxial film. X-ray diffraction revealed an almost perfect *c*-axis orientation of the films.

EXPERIMENT

The results shown were gained on a film which was partially untwinned revealing an anisotropy of the conductivity of approximately 15% at room temperature. Since the conductivity spectra along both directions turned out to be qualitatively similar, we present the results for $E \| b$, the most intensively studied orientation. Magnetic susceptibility measurement showed a sharp superconducting transition $[\Delta T(10\%-90\%) < 0.5\,\mathrm{K}]$ with an onset $T_C = 89.5$ K. Transmission experiments of the film-substrate system were performed utilizing a set of backward-wave oscillators.[19] The measurements were performed in a Mach-Zehnder interferometer arrangement which allows the measurements of transmission and phase shift. The optical parameters of the substrates were determined in separate experiments using the blank substrates. Utilizing the Fresnel formulas for a two-layer system the conductivity and the dielectric constant were determined from the observed spectra without assuming any particular model.

RESULTS AND DISCUSSION

Figure 1 shows the real part of the conductivity (lower panel) and the penetration depth (upper panel) as a function

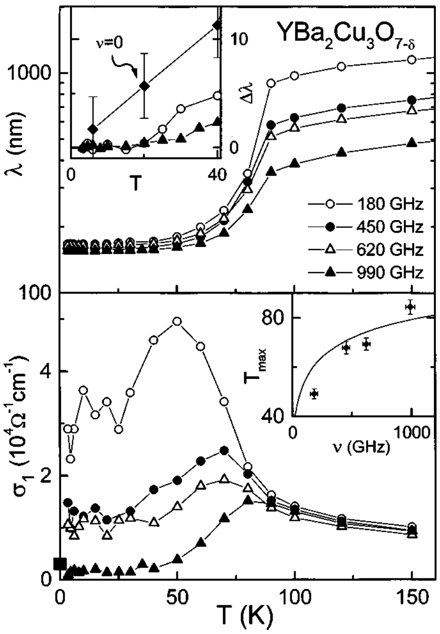

FIG. 1. Upper panel: Temperature dependence of the penetration depth. The inset shows the low-temperature behavior on a linear scale. The closed rhombs give a zero frequency extrapolation from the analysis of the complex conductivity (see text). Lower panel: Temperature dependence of the real part of conductivity at the same frequencies as in the upper panel. The closed square at $T=0$ K indicates the universal conductivity value (Ref. 4). The inset shows the temperature dependence of the conductivity peak maximums (rhombs) compared to the theoretical curve (solid line) (Ref. 20).

of temperature. The penetration depth is defined as $\lambda = c/\omega k$, with k being the imaginary part of the complex refraction index: $k = \mathrm{Im}[i(\sigma_1 + i\sigma_2)/\varepsilon_0\omega]^{1/2}$. The expression for λ gives the microwave result $\lambda = (\mu_0\omega\sigma_2)^{-1/2}$ in the limit $\sigma_1 \ll |\sigma_2|$ and the expression for the skin depth in the opposite limit $\sigma_1 \gg |\sigma_2|$. Below T_C the real part of the conductivity reveals a strong frequency dependence. The peak in the conductivity increases in height and is shifted to lower temperatures for decreasing frequencies. This behavior has been calculated in detail by Hirschfeld *et al.*[20] The inset shows the frequency dependence of temperature of the maximum conductivity, which qualitatively resembles the theoretical results.[20] We now focus on the low-temperature behavior of the conductivity to compare it with the result by P. A. Lee,[4] that the residual conductivity $\sigma_0(T \to 0, \nu \to 0)$ in a d-wave superconductor should be universal and independent of the impurity concentration, with $\sigma_0 = ne^2/(m\pi\Delta_0)$. Assuming an energy gap $\Delta_0 = 2.75k_BT_C$ and a penetration depth $\lambda_0 = 160$ nm, we estimate $\sigma_0 = 3\times10^3\ \Omega^{-1}\,\mathrm{cm}^{-1}$. This value is shown as a solid square in the lower panel of Fig. 1. At first sight our experimental results are not compatible with Lee's result.[4] However, it has been shown by Hirschfeld *et al.*[20] that in the Born limit this residual conductivity is effectively unobservable except at exponentially small temperatures. Instead, the conductivity tends to a higher value, determined by the concentration of impurities. $\sigma_1(\omega, T)$ as observed experimentally can only be explained if the theoretical predictions significantly different to the unitary limit[20] are used.

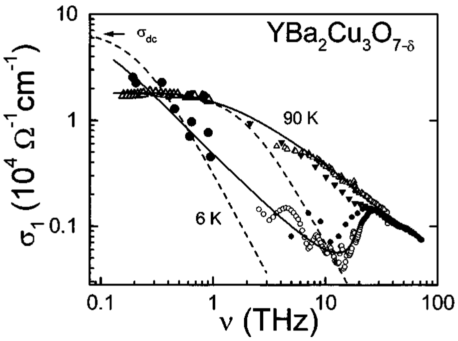

FIG. 2. Frequency dependence of the real part of conductivity at temperatures just above T_C (90 K, triangles) and far below T_C (6 K, circles) compared to the infrared data. The IR data ($\nu > 2$ THz) are taken from Ref. 21 (closed symbols) and Ref. 22 (open symbols). Solid lines are drawn to guide the eye. Dashed lines represent a Drude fit (see text).

The upper panel of Fig. 1 shows the temperature dependence of the penetration depth. The penetration depths reveal a sharp drop at T_C for all frequencies and extrapolates towards values of 160 nm at the lowest temperatures. The low-temperature behavior of the penetration depth is shown in the inset of the upper panel of Fig. 1. According to the recent theoretical models[20] a linear temperature dependence of the penetration depth of a d-wave superconductor can only be expected at zero frequencies. With increasing microwave frequency the temperature dependence of the penetration depth strongly becomes reduced,[20] a behavior which indeed is observed.

Figure 2 shows the real part of the conductivity as a function of frequency for two temperatures. To demonstrate the sample-independence of the conductivity we took single crystal[21] and thin film data[22] for comparison. The IR data are plotted on absolute scale without any arbitrary shifts. The data clearly reveal a Drude-like behavior at 90 K with a relaxation rate $\Gamma = (2\pi\tau)^{-1}$ of the order of 3 THz. The data also provide clear experimental evidence that a narrow conductivity peak develops at the lowest temperatures ($T < T_C/10$). The dashed lines represents a Drude fit to the data, using a σ_{dc} value that will be explained later. The solid lines are mere interpolations between the submillimeter and the FIR results. The discrepancies between the dashed and the solid lines at $T=6$ K may be due to the fact that the very low-frequency FIR spectra are ill-defined only. In a range from 300 GHz to 30 THz a strong suppression of the conductivity becomes apparent revealing a gaplike feature around approximately 10 THz. This characteristic frequency $\nu^* \approx 10$ THz most probably corresponds to a crossover of the elastic and inelastic scattering rates.[23] The conductivity in the superconducting phase reveals a peak close to 23 THz ≈ 1100 K. If we assign this value to $4\Delta_0$,[23] where Δ_0 is the maximum gap value, we conclude that $2\Delta_0 \approx 6.4kT_C$.

The most relevant results are shown in Fig. 3. Here we show the real (lower panel) and imaginary part (upper panel) of the conductivity as a function of frequency for different temperatures. Following suggestions of Jiang *et al.*[24] and Schachinger *et al.*[25] we have plotted the product $\sigma_2 \cdot \nu$. The reason to present data in this way is that the square of the penetration depth is directly related to this quantity through

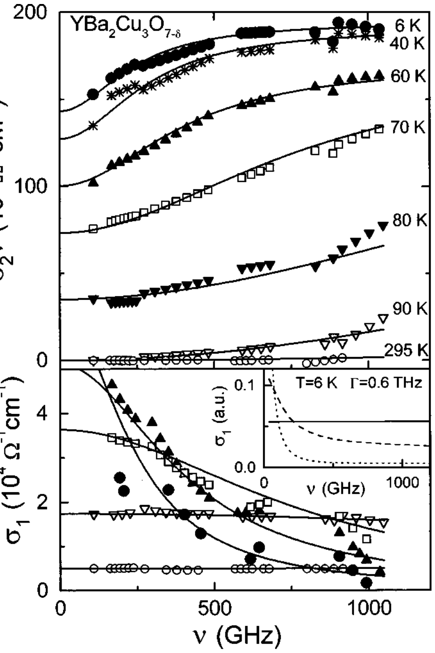

FIG. 3. Frequency dependence of the complex conductivity of $YBa_2Cu_3O_{7-\delta}$ film at different temperatures. Upper panel: the product $\sigma_2 \cdot \nu$; lower panel: σ_1. The lines are fits according to Eq. (1). The insert demonstrates the effect of varying the effective scattering cross section σ within a d-wave model of superconductivity (Ref. 26). The curves are calculated assuming $\Delta(0)=3.8$ THz, $T_C=90$ K, and $\sigma=1$ (unitary limit, solid line), $\sigma=0.7$ (dashed) and $\sigma=0$ (Born limit, dotted).

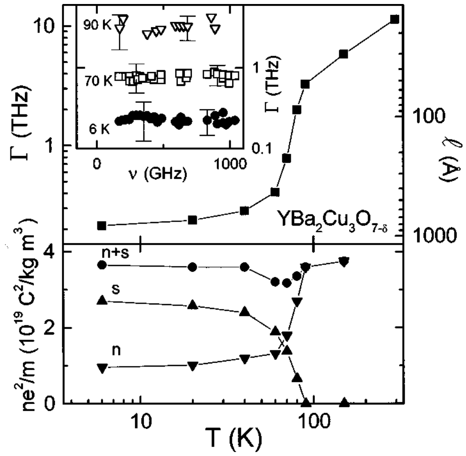

FIG. 4. Temperature dependence of the quasiparticle scattering rate $\Gamma=(2\pi\tau)^{-1}$ (upper panel) and spectral weight (lower panel) as extracted from the fits to the data of Fig. 3. The right-hand side of the upper panel gives the mean free path, calculated as $l=v_F\cdot\tau$, with $v_F=1100$ km/s being the Fermi velocity (Ref. 26). The inset shows the frequency dependence of the scattering rate. The data in the lower panel give the spectral weight of the normal (n) and the superconducting (s) contributions and the sum of both components ($n+s$).

$\lambda^{-2}(0)\sim\lim_{\nu\to 0}(\sigma_2\nu)$. With the decreasing temperature a well-defined minimum around $\nu\sim 0$ is observed in $\sigma_2\cdot\nu$ (upper part of Fig. 3) and a Drude-like peak centered at zero frequency develops in the superconducting state in σ_1 (lower part of Fig. 3). This feature directly reflects the electromagnetic response of the quasiparticles, which do not contribute to the superconducting condensate. Analyzing the amplitude and the width of this process we can extract the information about the spectral weight and the scattering rate of the quasiparticles. The real and the imaginary part of the conductivity were fitted simultaneously assuming a Drude-like response of the quasiparticles in the superconducting state with a temperature-dependent, but frequency-independent scattering rate. The electromagnetic properties of the superconducting condensate were also taken into account adding the appropriate σ_s^* term to the Drude conductivity. The solid lines in Fig. 3 were fitted using

$$\sigma_1+i\sigma_2=\sigma_D^*+\sigma_s^*=\frac{n_n e^2}{m}(\tau^{-1}-i\omega)^{-1}+\frac{n_s e^2}{m}[\pi\delta(0)/2+i/\omega], \quad (1)$$

where the spectral weights of the normal and the superconducting components n_n, n_s, and the scattering rate τ^{-1} were treated as adjustable parameters. Only for temperatures $T \geqslant 200$ K n_n and n_s were kept constant due to the low accuracy of $\sigma_2(\nu)$. The agreement between model calculations and experimental results is quite satisfactory.

The insert in Fig. 3 demonstrates the effect of varying the effective scattering cross section within a weak-coupling calculation with finite scattering rate assuming d-wave pairing.[26] The line corresponding to the unitary limit shows no frequency dependence in the frequency range below 1 THz and at $T=6$ K. Already the scattering cross section $\sigma=0.7$ has a significant frequency dependence at this temperature and can qualitatively explain the observed conductivity spectra.

Figure 4 shows the temperature dependence of the fit parameters obtained using Eq. (1). The scattering rate collapses from a value close to 3 THz at T_C to 0.2 GHz at 40 K and then remains almost constant for further decreasing temperatures. Most probably and in accord with many other experimental findings, a gap develops in the spin-fluctuation spectrum at the superconducting phase transition temperature making the quasiparticles long-lived below T_C. From the upper panel of Fig. 4 we can conclude that for temperatures $T>40$ K the scattering rates are determined mainly by inelastic scattering processes from spin fluctuations. Below 40 K elastic impurity scattering processes dominate.

The lower part of Fig. 4 shows the superconducting and "normal" conducting spectral weights extracted from the fits. Even for $T<T_C/10$, the quasiparticle spectral weight remains at a value of 1/4 as compared to the normal state. This fact agrees well with theoretical estimations,[18,23] which predict a finite concentration of the nonpaired quasiparticles in the presence of impurities even at $T=0$. In addition, we show the temperature dependence of the n_s+n_n. It is a remarkable and important result that this value remains almost constant. Since the normal-state conductivity spectrum is much broader than the simple Drude process, the analysis based on Eq. (1) extracts only the spectral weight of the single Drude at zero frequency. This spectral weight is given by the area under the $T=90$ K Drude curve in Fig. 2. Thus,

only the spectral weight under this curve is transferred into the superconducting condensate.

The fits to the experimental results, shown in Fig. 3, allow the determination of the zero frequency penetration depth λ_0. The resulting low-frequency values are plotted in the inset of the upper panel of Fig. 1. The only three points that have been measured reveal a linear dependence of the penetration depth with a slope $d\lambda/dT \approx 3$ Å/K.

Finally, assuming that the quasiparticle relaxation time can be frequency dependent, we tried to extract $\tau(T,\nu)$ from our data. In order not to mix the effects of the normal and the superconducting component, the term σ_s^* describing the superconducting condensate has been subtracted from the experimental conductivity. Now the frequency dependence of the scattering rate follows from[24] $\tau^{-1}(\omega)=\omega\sigma_1^n/\sigma_2^n$, where the notation σ^n indicates the normal conducting fraction. The frequency dependence of τ is shown as inset in Fig. 4 and reveals that a frequency-independent scattering rate is a good approximation in the frequency range of the present experiment.

CONCLUSIONS

In conclusion, we have measured the real and imaginary parts of conductivity of partly untwinned $YBa_2Cu_3O_{7-\delta}$ films in the frequency range $0.1<\nu<1$ THz. The analysis of the experimental spectra allows the determination of spectral weight and scattering rates of the quasiparticles directly. From the temperature dependence of the conductivity we conclude that the impurity scattering cannot be described within the unitary limit. The penetration depth, which is expected to depend linearly on temperature in the zero-frequency limit in a *d*-wave superconductor, is strongly suppressed at submillimeter frequencies.

ACKNOWLEDGMENTS

We acknowledge stimulating discussions with A. Kampf, P. J. Hirschfeld, D. van der Marel, and D. Rainer. This work was supported by the BMBF via Contract No. 13N6917/0—Elektronische Korrelationen und Magnetismus.

[1] D. A. Bonn and W. N. Hardy, in *Physical Properties of High Temperature Superconductors V*, edited by D. M. Ginsberg (World Scientific, Singapore, 1996), p. 7.
[2] D. B. Tanner and T. Timusk, in *Physical Properties of High Temperature Superconductors III*, edited by D. M. Ginsberg (World Scientific, Singapore, 1992), p. 363.
[3] S. Tajima, Supercond. Rev. **2**, 125 (1997).
[4] P. A. Lee, Phys. Rev. Lett. **71**, 1887 (1993).
[5] K. Holczer, L. Forro, L. Mihaly, and G. Grüner, Phys. Rev. Lett. **67**, 152 (1991).
[6] D. A. Bonn, P. Dosanjh, R. Liang, and W. N. Hardy, Phys. Rev. Lett. **68**, 2390 (1992).
[7] F. Gao, J. W. Kruse, C. E. Platt, M. Feng, and M. V. Klein, Appl. Phys. Lett. **63**, 2274 (1993).
[8] D. A. Bonn, S. Kamal, K. Zhang, R. Liang, D. J. Baar, E. Klein, and W. N. Hardy, Phys. Rev. B **50**, 4051 (1994).
[9] H. Srikanth, B. A. Willemsen, T. Jakobs, S. Sridhar, A. Erb, E. Walker, and R. Flükiger, Phys. Rev. B **55**, 14 733 (1997).
[10] S. Hensen, G. Müller, C. T. Rieck, and K. Scharnberg, Phys. Rev. B **56**, 6237 (1997).
[11] M. C. Nuss, P. M. Mankiewich, M. L. O'Malley, E. H. Westerwick, and P. B. Littlewood, Phys. Rev. Lett. **66**, 3305 (1991).
[12] I. Francois, C. Jaekel, G. Kyas, D. Dierickx, O. Van der Biest, R. M. Heeres, V. V. Moshchalkov, Y. Bruynseraede, H. G. Roskos, G. Borghs, and H. Kurz, Phys. Rev. B **53**, 12 502 (1996).
[13] A. Frenkel, F. Gao, Y. Liu, J. F. Whitaker, C. Uher, S. Y. Hou, and J. M. Phillips, Phys. Rev. B **54**, 1355 (1996).
[14] Ch. Ludwig, Q. Jiang, J. Kuhl, and J. Zegenhagen, Physica C **269**, 249 (1996).
[15] U. Dähne, T. Amrein, and Y. Goncharov, Physica C **235-240**, 2066 (1994); U. Dähne Y. Goncharov, N. Klein, N. Tellmann, G. Kozlov, and K. Urban, J. Supercond. **8**, 129 (1995).
[16] L. A. de Vaulchier, J. P. Vieren, Y. Guldner, N. Bontemps, R. Combescot, Y. Lemaître, and J. C. Mage, Europhys. Lett. **33**, 153 (1996); S. Djordjevic, L. A. de Vaulchier, N. Bontemps, J. P. Vieren, Y. Guldner, S. Moffat, J. Preston, X. Castel, M. Guilloux-Viry, and A. Perrin, Eur. Phys. J. B **5**, 847 (1998).
[17] J. Annett, N. Goldenfeld, and S. R. Renn, Phys. Rev. B **43**, 2778 (1991).
[18] P. J. Hirschfeld and N. Goldenfeld, Phys. Rev. B **48**, 4219 (1993).
[19] A. A. Volkov, Yu. G. Goncharov, G. V. Kozlov, S. P. Lebedev, and A. M. Prochorov, Infrared Phys. **25**, 369 (1985).
[20] P. J. Hirschfeld, W. O. Putikka, and D. J. Scalapino, Phys. Rev. B **50**, 10 250 (1994); P. J. Hirschfeld, W. O. Putikka, and D. J. Scalapino, Phys. Rev. Lett. **71**, 3705 (1993).
[21] D. N. Basov, R. Liang, B. Dabrowski, D. A. Bonn, W. N. Hardy, and T. Timusk, Phys. Rev. Lett. **77**, 4090 (1996).
[22] J. Schützmann, W. Ose, J. Keller, K. F. Renk, B. Roas, L. Schultz, and G. Saemann-Ischenko, Europhys. Lett. **8**, 679 (1989).
[23] S. M. Quinlan, P. J. Hirschfeld, and D. J. Scalapino, Phys. Rev. B **53**, 8575 (1996).
[24] C. Jiang, E. Schachinger, J. P. Carbotte, D. Basov, and D. Timusk, Phys. Rev. B **54**, 1264 (1996).
[25] E. Schachinger, J. P. Carbotte, and F. Marsiglio, Phys. Rev. B **56**, 2738 (1997).
[26] M. J. Graf, M. Palumbo, and D. Rainer, Phys. Rev. B **52**, 10 588 (1995).