Journal of Alloys and Compounds, 195 (1993) 663–666
JALCOM 7553

Optical conductivity and carrier relaxation rate in the normal state of high T_c thin films.

A. EL AZRAK[a], R. NAHOUM[a], A.C. BOCCARA[a], N. BONTEMPS[b], M. GUILLOUX-VIRY[c], C. THIVET[c], A.PERRIN[c], Z.Z. LI[d], H. RAFFY[d]

[a] Optique Physique (UPR CNRS 0005) ESPCI, 10 rue Vauquelin 75231 Paris Cedex 05 (France)

[b] LPMC (URA CNRS 1437) ENS, 24 Rue Lhomond, 75231 Paris Cedex 05 (France)

[c] LCSIM (URA CNRS 1495), Université de RENNES I, 35042 Rennes Cedex (France)

[d] Physique des Solides, (LA CNRS 2) Université Paris-Sud, 91405 Orsay Cedex (France)

Abstract

We report the optical conductivity of $YBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$ thin films at room temperature. The optical conductivity in the 500-7000 cm^{-1} frequency range is analysed in terms of a frequency dependent relaxation time τ and effective mass of the normal carriers. The relaxation rate $1/\tau$ exhibits the same linear dependence on frequency whatever the sample. The assumption that $1/\tau$ also depends linearly on temperature accounts satisfactorily for the temperature variation (90-300K) of the transmission in the 1000-7000 cm^{-1} range. These results are consistent with the marginal Fermi liquid model.

I. Introduction

There is presently large experimental evidences in $La_{2-x}Sr_xCuO_4$ [1], $YBa_2Cu_3O_{6+x}$ [2,3,4], $Nd_{1+y}Ba_{2-y}Cu_3O_{6+x}$ [4] $Bi_2Sr_2CaCu_2O_{8+\delta}$ [5] that free carriers within the conductive CuO_2 planes are responsible for the infrared reflectivity spectrum up to 1 eV (8000 cm^{-1}). Although still controversial, a generalised Drude conductivity where both the effective mass m* and the relaxation time τ of the normal state carriers are frequency dependent has been invoked as a description of the infrared response [6,7,8]. The relaxation rate $1/\tau$ was found to exhibit a linear variation with frequency in a large frequency domain, up to 2000 cm^{-1} on a single domain $YBa_2Cu_3O_7$ crystal [8], and even up to 6000 cm^{-1} on Bi based single crystals [9].

We present in this paper infrared conductivity data of $YBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$ thin films derived from reflectivity spectra extending up to 25 000 cm^{-1} [10]. We find that the linear variation of the relaxation rate $1/\tau$ versus frequency extends up to 6000 cm^{-1} and that $1/\tau$ is the same *for all the compounds studied*. We find that in the 1000-6000 cm^{-1} range, the temperature change of the transmission between 90 K and 300 K can only be quantitatively accounted for by a linear variation of the relaxation rate with temperature. As a whole, our findings are consistent with the marginal Fermi liquid (MFL) model [11].

II. Experimental

We have studied the following samples :

$YBa_2Cu_3O_7$ (3000Å) on MgO
$YBa_2Cu_3O_7$(1000Å) on $SrTiO_3$
$GdBa_2Cu_3O_7$ (2000Å) on MgO
$Bi_2Sr_2Ca_1Cu_2O_8$ (3500Å) on MgO

The preparation of the $YBa_2Cu_3O_7$ thin films has been reported elsewhere [12].They were epitaxially grown in-situ by laser ablation i.e. the axes of the films are aligned with those of the substrate. They present narrow transitions (ΔT_c<0.5 K) with T_c(R=0) in the range 85.2-86.5 K measured during fast cooling. T_c for the Gd sample is slightly smaller: T_c(R=0)=79.5 K [12].

The $Bi_2Sr_2CaCu_2O_{8+\delta}$ thin film was epitaxially grown in situ by RF magnetron single target sputtering on an MgO substrate kept at high temperature (~735°C) [13]. The critical temperature T_c(R=0) is 80K.

All samples are highly c-axis oriented [12,13].

0925-8388/93/$06.00

Reflectivity near normal incidence and transmission spectra have been recorded in the 500-7000 cm^{-1} spectral range with an IFS66 Bruker Fourier-transform spectrometer (Transmission data only start at 1000 cm^{-1} due to the MgO absorption) and in the 5000-25000 cm^{-1} with a Cary 17 spectrophotometer.

Kramers-Krönig (KK) analysis from reflectivity data has been used in order to establish the real part of the conductivity for those samples which are thick enough so that the substrate plays no role in the reflected flux. Details of the KK procedure have been given elsewhere [10].

In the 1000Å film, the substrate gives rise to extra contributions to the conductivity derived through the KK procedure : the results on this sample make clear the experimental limits of the KK analysis. These deviations appear between 1000 and 2000 cm^{-1}, and are assigned to the substrate absorption which is most effective below this range [14].

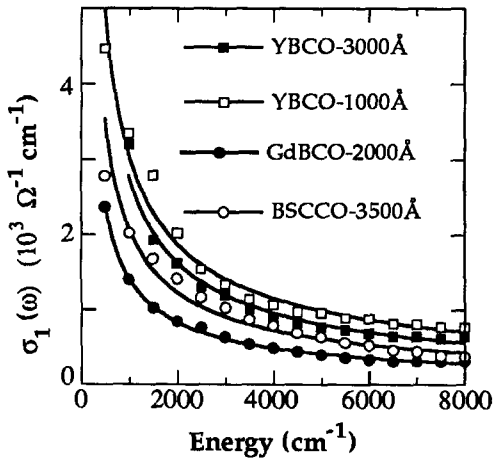

Figure1 . Real part of the conductivity . The solid lines show the best fit to a power law.

Fig.1 shows the real part of the conductivity in the range 500-8000 cm^{-1}, for all our samples .

We have measured the transmission spectrum of several thin films $YBa_2Cu_3O_7/MgO$ for various thicknesses between 300 K and 100 K in the range 1000-7000 cm^{-1}. We report on fig.3 only the results of the 3000Å thick sample, which has been studied also by reflectivity.

III - Discussion

3.1. Frequency dependence of the conductivity

In order to characterize the non-Drude behavior, we fitted the conductivity with a $\omega^{-\alpha}$ power law; this is shown on Fig.1. The agreement is satisfactory, the thinnest film (1000Å) exhibiting some deviations to the power law as mentioned above. The striking result is that we find an exponent which is the same for all the films, namely 0.77±0.05.

3.2. Frequency dependence of the relaxation rate

This non-Drude behavior can be further analysed, as mentioned in the introduction, in terms of a frequency dependent relaxation rate $1/\tau$ and effective mass [6,7,8].

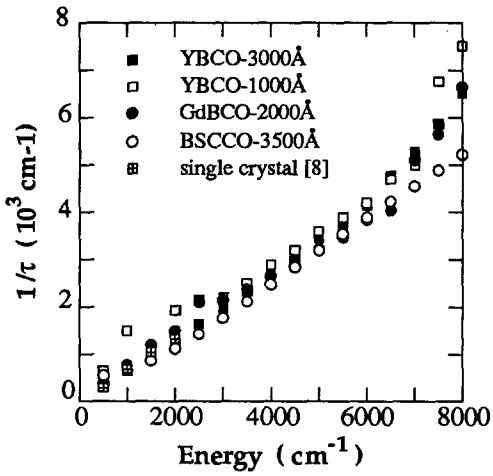

Figure 2 . Relaxation rate $1/\tau$ of the carriers from thin films (this study) and an untwinned crystal [8].

We show the frequency dependence of the relaxation rate in fig. 2. The remarkable result here is that the relaxation rate exhibits a linear dependence which is the same for all the samples up to 6000 cm^{-1}. A slight discrepancy appears for the thinnest sample at 1000-2000 cm^{-1}. Above 6000 cm^{-1}, the points start to depart from the straight line. This is presumably due to contributions to the conductivity which are not related to the carriers.

We have also reported on fig.2 the points extracted under similar assumptions from the conductivity σ_a of an untwinned crystal (the electric field being polarized

parallel to the a axis) hence presumably associated only with the conductivity within the CuO_2 planes[8] . These points fall right on top of ours. This suggests strongly that the chain contribution is not seen in the films, which is also confirmed by the relaxation rate of the Bi based compound (no chains) exhibiting the same behavior.

Besides, the fact that the frequency dependence of the conductivity is the same for all films also suggests a common physical origin for the anomalous behavior of the infrared conductivity in all compounds, hence ruling out the chains which are specific of the 123 material.

This linear dependence of the relaxation rate is found experimentally to be:

$$1/\tau = (0.67\pm0.07)\ h\nu \quad (1)$$

There are presently several theories for cuprate superconductors which investigate the optical conductivity and predict an anomalous frequency dependence of the optical conductivity or a relaxation rate which varies linearly in frequency (or both).

One is the marginal Fermi liquid (MFL), where one expects [11]:

$$1/\tau_{MFL} = 2\ \pi\ \lambda\ (h\nu+\pi k_B T) \quad (2)$$

where λ is a coupling constant. From eq.1, $\lambda=0.11\pm0.01$

This linear dependence is expected to hold up to a cut-off frequency ν_c related to the energy spectrum of the bosonic spectrum which is flat up to this frequency. The linearity domain which we find to extend up to 6000 cm^{-1} appears unusually large if refering to this prescription.

In order to make a more quantitative comparison with the frequency dependence of the conductivity in this model, one has to consider that the frequency range extends below and beyond ν_c. We have analysed the computed conductivity of ref.11 and identified the exponent α : we find 0.74 for the conductivity computed at 300K, in surprisingly good agreement with what we find on our films.

3.3. Temperature dependence

Eq.2 thus implies a temperature dependence of the relaxation rate associated with its frequency dependence. Since λ is determined from eq.1, the temperature dependence of $1/\tau$ is unambiguously determined. It should give rise to a temperature variation of the reflectivity or of the transmission in the mid-infrared range. A small although measurable (~3% between 250 and 105 K) variation of the reflectivity has indeed been observed [7].

We have ourselves measured the temperature variation of the transmission spectrum in the relevant frequency range, namely 1000-7000 cm^{-1}, for the $YBa_2Cu_3O_7$ (3000Å)/MgO film. Fig.3 shows the ratio of the transmission spectrum of the film to the transmission of the MgO substrate measured by switching "in situ" from the sample to the reference substrate. We take this ratio in view of cancelling as best as possible the interference pattern due to the cryostat windows. We have also measured independently the variation of the transmission of the substrate and found it negligible between 2000 and 8000 cm^{-1}.

When the temperature decreases, we observe a somewhat uniform decrease of the transmission, a result consistent with reflectivity data, which show a uniform increase [7]. For sake of clarity, we only show on fig.3 the spectra at two temperatures. Although the overall transmission is very small (~1%), the relative change with temperature is much larger than in reflectivity.

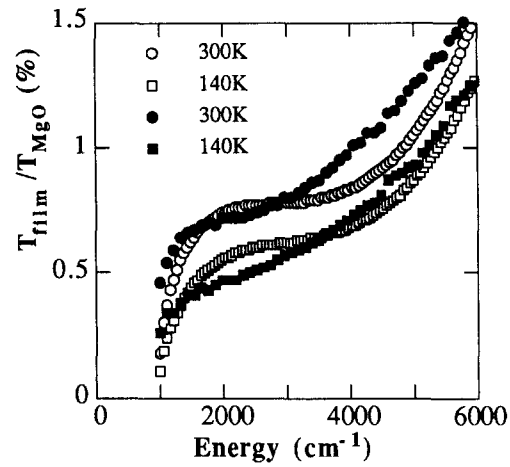

Figure 3. Experimental (full symbols) and computed (open symbols) transmission of a YBCO-2900Å film

We now compare the experimental spectra with computed ones. The calculation uses for the relaxation rate eq.2, with $\lambda=0.11$, and takes into account the possible interference patterns that may occur within the film (2950±50Å) and the substrate (500 μm), and

assumes a temperature independent f-sum rule. The associated plasma frequency is ω_p=23 300 cm^{-1} from room temperature reflectivity data. We thus get computed spectra with no adjustable parameters. The result of this comparison is shown on fig.3.

The change with temperature of the computed spectra compares satisfactorily with the experimental one. The absolute magnitude of the transmission is right taking 2900 Å for the thickness of the film; this is consistent with the accuracy within which the thickness is known.

Finallly, we mention that other temperature dependences which could be consistent with fig.2, e.g. :

$$1/\tau = \mu \sqrt{(\pi^2 k_B^2 T^2 + h^2 \nu^2)} \quad (3)$$

must be ruled out because they yield too small a temperature change in the high energy part of the transmission spectrum.

IV - Conclusion

There are presently several theoretical models (the marginal Fermi liquid [11], the nested Fermi liquid [15], gauge theories [16]) which yield very similar predictions as far as the optical conductivity and the relaxation rate are concerned. It is therefore a delicate question to build up reliable comparisons between theory and experiment.

All our samples exhibit a dependence of the optical conductivity which is well described by a power law with a single exponent (-0.77), up to 6000 cm^{-1}. This behavior may be analysed in terms of a relaxation rate which varies linearly with frequency up to a very large energy (6000 cm^{-1}). Accordingly, a linear variation of the relaxation rate wih temperature accounts satisfactorily for the observed temperature dependence of the transmission spectrum. These results appear in quantitative agreement with the marginal Fermi liquid model.

Acknowledgements

We are very much indebted to Pr C. VARMA, Pr R. COMBESCOT, Pr M. GABAY and Pr P. LEDERER for fruitful suggestions .

References

1. S. Uchida, T. Ido, H. Takagi, T. Arima, Y. Tokura and S. Tajima, Phys. Rev. B 43 (1991) 11237
2. J. Orenstein, G.A. Thomas, A.J. Millis, S.L. Cooper, D.H. Rapkine,T. Timusk, L.Schneemeyer and J.V. Warszczak, Phys. Rev. B 42, (1990) 6342
3. G.A. Thomas, S.L. Cooper, J. Orenstein, D.H. Rapkine, A.J. Millis, J.V. Waszczak and L.F. Schneemeyer, Super. Sci. Technol. 4 (1991) S235
4. J. Bouvier, N. Bontemps, M. Gabay, M. Nanot and F. Queyroux, Phys. Rev. B 45 (1992) 8065
5. J.H. Kim, I. Bozovic, D.B. Mitzi, A. Kapitulnik and J.S. Harris Jr, Phys. Rev. B 41 (1990) 7251
6. G.A. Thomas, J. Orenstein, D.H. Rapkine, M. Capizzi, A.J. Millis, R.N. Bhatt, L.F. Schneemeyer and J. Warszczak, Phys. Rev. Lett. 61 (1988) 1313
7. Z. Schlesinger, R.T. Collins, F. Holtzberg, C. Feidl, G. Koren and A. Gupta, Phys. Rev. B 41 (1990) 11237
8. Z. Schlesinger, R.T. Collins, F. Holtzberg, C. Feild , S.H. Blanton,U. Welp, G.W. Crabtree, Y. Fang and J.Z. Liu, Phys. Rev. Lett. 65 (1990) 801
9. I. Terasaki, S. Takebayashi, I. Tsukada, A. Maeda and K. Uchinokura, Physica C 185-189 (1991) 1017
10. A. El Azrak, R. Nahoum N. Bontemps, M. Guilloux-Viry, C. Thivet, A.Perrin, S. Labdi, Z.Z. Li and H. Raffy (submitted for publication)
11. P.B. Littlewood and C.M. Varma J. Appl. Phys. 69 (1991) 4979
12. M.G. Karkut, M. Guilloux-Viry, A. Perrin, J. Padiou and M. Sergent, Physica C 179 (1991) 262
13. Z.Z. Li, S. Labdi, A. Vaures, S. Megtert and H. Raffy , Proceedings of ICAM-91 EMRS, Conf., Strasbourg 1991, Ed. L .Correra , Elsevier Sc. Pub (1992) 487
14. M. Sato, K. Nagasaka, M. Ihara and T. Kimura, J. Phys. Soc. Jap. 60 (1991) 4337
15. J. Ruvalds and A. Virosztek, Phys. Rev. B 43 (1991) 5498
16. L.B. Ioffe and G. Kotliar Phys. Rev. B 42 (1990) 10348