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REFLECTIVITY AND TRANSMISSION SPECTRA OF $Bi_2Sr_2Ca_1Cu_2O_8$ SPUTTERED THIN FILMS

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We report on the room-temperature reflectivity and transmission spectra of $Bi_2Sr_2Ca_1Cu_2O_8$ thin films. By fitting simultaneously both spectra, we determine the plasma frequency and characterize several oscillators in the infrared and visible range. The role of the mid infrared band is emphasized.

1.INTRODUCTION

Optical reflectivity on high T_c superconductors is among the basic techniques which yields parameters of fundamental interest for these materials, such as the plasma frequency ω_p (hence the density of the carriers in the normal state), or possibly the superconducting gap. Transmission experiments being restricted to thin films, are much less popular. The largest amount of experimental work deals with $YBa_2Cu_3O_{6+x}$[1], and much less with the $Bi_2Sr_2Ca_1Cu_2O_8$ compound.

We report in this work room temperature reflectivity and transmission measurements in a large spectral range (400-27000 cm^{-1}) of two $Bi_2Sr_2Ca_1Cu_2O_8$ thin films on MgO substrate having different characteristics in particular their transition temperature and their dc resistivity. We find slightly different spectra that we can fit however with similar parameters. The reflectivity and transmission spectra cannot be simultaneously accounted for with a single Drude contribution extending up to 10000 cm^{-1}, whereas this interpretation cannot usually be ruled out when only the reflectivity spectrum are available[2]. Indeed, in earlier work, the non Drude-like behavior in the infrared range (up to 10000 cm^{-1}) was evidenced through variable temperature measurements[1, 3].

This paper is aimed at the determination of the plasma frequency ω_p defined by :

$$\omega_p = (4\pi ne^2/m^*)^{1/2} \qquad (1)$$

where n is the density of the free carriers and m* is the carrier effective mass. The values we find in both films (around 9000 cm^{-1}) though

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smaller, are not far from earlier determinations[3,4,5].

We find large room temperature relaxation rates (1300-2000 cm^{-1}) compared to single crystal data[3] (69 cm^{-1}). They actually relate to the large dc resistivities measured on these films. Finally, the localisation of higher energy (visible) bands is made considerably easier by the transmission data.

2.FILM PREPARATION AND CHARACTERISATION

The thin films have been prepared by a two step process[6,7]. Amorphous deposits were obtained on (100) oriented MgO single crystal substrates by d.c. triode sputtering from a single composite oxide target. They were subsequently annealed at 820°C in flowing O_2-N_2. The composition of annealed deposits is close to that of the target.
X-ray diffractions studies show that the samples are essentially composed of the 2212 phase and are highly textured with copper-oxide planes parallel to the substrate (see fig. 3 of Ref. 2). Before performing optical studies, the superconducting transition temperatures have been measured resistively, using four-point spring-loaded contacts in order not to contaminate the surface. The samples are very stable (no change in T_c after several months).

3.EXPERIMENTAL

Transmission and near normal incidence (15°) reflectivity spectra are recorded with an IFS66 Brucker Fourier-Transform spectrophotometer in the 400-7000 cm^{-1} range, and a Cary 17 spectrophotometer between 5000 and 27000 cm^{-1}. We use as a reference an Ag mirror.

4.RESULTS

Figures 1 and 2 show the reflectivity and transmission spectra of sample S1. Figure 1a shows the entire spectrum up to 25000 cm^{-1}, figure 1b displays an expanded view of the reflectivity in the 400-1700 cm^{-1} range. Figures 3 and 4 show the same spectra for sample S2.
The infrared (IR) reflectivity of the two films under investigation is comparable but somewhat lower than the one observed on single crystals[2,3], and it is definitely higher than the one reported Pb-doped sputtered films[8].

Both samples exhibit at 670 cm^{-1} a dip in the spectrum which may be compared to a similar feature at the same frequency however on $Bi_2Sr_2Ca_1Cu_2O_8$ pellets[9] and is also mentioned though not explicitly shown, on a single crystal[10].

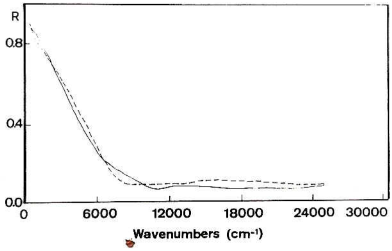

Figure 1a

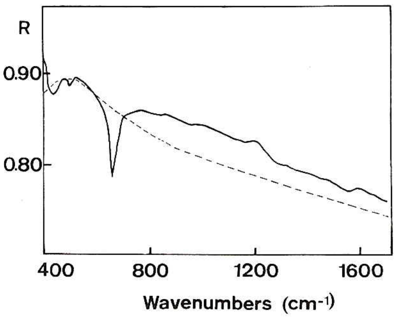

Figure 1b

Figure 1

Reflectivity spectrum of sample S1

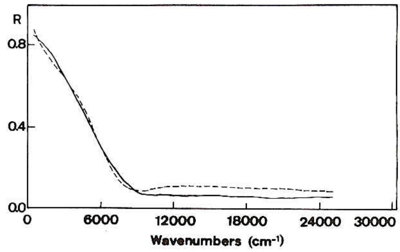

Figure 3a

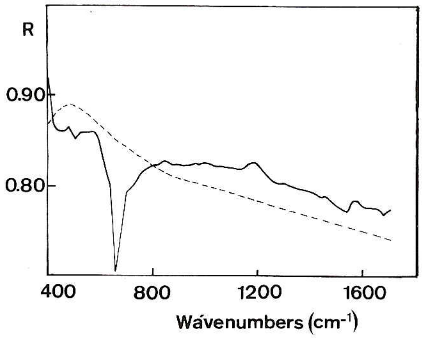

Figure 3b

Figure 3

Reflectivity spectrum of sample 2

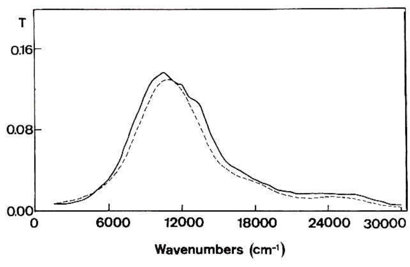

Figure 2

Transmission spectrum of sample S1

(———) : experimental curve
(-----) : fitted curve

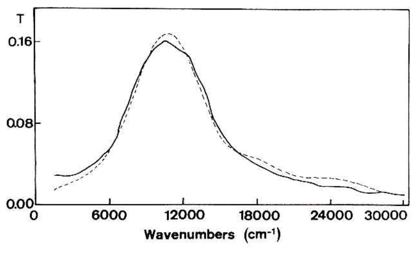

Figure 4

Transmission spectrum of sample S2

(———) : experimental curve
(-----) : fitted curve

Transitions occuring in the 1000-25000 cm^{-1} energy range are barely detectable[11], although weak structures seem to be present at approximately 14000 and 20000 cm^{-1}.

The transmission spectra shown on fig. 2 and 4 are obtained by dividing the raw spectra by the transmission spectrum of the MgO. The low transmission below 3000 cm^{-1}, steadily increasing up to 10000 cm^{-1} is the counterpart of the high IR reflectivity in the same spectral range. The occurence of absorption bands is fully ascertained by the decrease of the transmission and the structure of the spectra, which suggest three bands around 14000, 21000 and 29000 cm^{-1}.

5.ANALYSIS AND DISCUSSION

We have chosen to model the reflectivity and transmission spectra by including the following contributions :

- A conventional Drude term, associated with a plasma frequency ω_p and a relaxation rate γ which does not depend upon the frequency.
- An oscillator in the infrared (580 cm^{-1}).
- A "mid infrared band" defined by its resonance frequency ω_2, its width γ_2 and its oscillator strength ω_{f2}.
- 3 transistions in the visible at 15000, 21000 and 30000 cm^{-1}.

We write the dielectric constant as follows :

$$\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_p^2}{\omega\,(\omega + i\,\gamma)} + \frac{\omega_{fk}^2}{\omega_k^2 - \omega^2 - i\,\omega\,\gamma_k} \qquad (2)$$

where the ω_k, γ_k and ω_{fk} are respectively the frequency, width and oscillator strength of the k^{th} oscillator. ε_∞ is the dielectric constant in the limit of very high frequency and is associated with an average refractive index n. We have obtained an estimate of the value of n, by looking at the minimum of reflected flux at 15800 cm^{-1}, hence in the range where the absorption is the smallest (see fig. 2 and 4). This minimum of reflection occurs, when absorption is negligible, at the Brewster incidence, $\tanh(I_b)=n=(\varepsilon(\omega))^{1/2}$. We have thus deduced $\varepsilon_\infty \approx 3$.

From the generalised dielectric constant given by (2), we define the index of refraction of the medium N, which is the relevant quantity in order to compute the reflectivity or the transmission :

$$N = (\varepsilon(\omega))^{1/2} = n - ik \qquad (3)$$

Our films being thick enough to absorb more than ≈ 80 %, we have used a simplified two waves interference model instead of a multiple one[5]. Within the substrate, we have neglected possible interference effects considering that it is thick (600 microns) compared to the wavelength and presumably its flatness is not well defined enough so that it is unlikely that the waves exhibit phase coherence. We have thus only added the energies of reflected or transmitted rays,restricting here the calculation to the first two waves due to weak interface reflections.

We thus write the reflectivity R and transmission T as follows :

$$R = |r_{01}+t_{01}t_1r_{12}t_1t_{10}|^2 + |t_{01}t_1t_{12}t_2r_{20}t_2t_{21}t_1t_{10}|^2 \quad (4)$$

$$T = |t_{01}t_1t_{12}|^2 \, [\,|t_2t_{20}|^2 + |t_2r_{20}t_2r_{21}t_2t_{20}|^2] \quad (5)$$

The indices 0, 1 and 2 specify the air, the film and the substrate respectively N_j is the index of refraction of the j^{th} medium. The quantities r_{jk}, t_{jk} and t_j are defined as :

$$r_{jk} = (N_j - N_k)/(N_j + N_k) \quad (6)$$

$$t_{jk} = (2N_j)/(N_j + N_k) \quad (7)$$

$$t_j = \exp(-2i\pi k_j e_j/\lambda) \quad (8)$$

e_j is the thickness of the j^{th} medium.

Since the reflectivity and transmission of the substrate have been recorded separately, we extract the index of refraction N_2 from these data. Therefore, the only adjustable parameters in eq. (4) and (5) are related to the film.

Our best fits are shown in figures 1, 2, 3 and 4. We show in table I the parameters that we have thus determined for both samples. The quality of the fit is good in the infrared range but the computed reflectivity spectrum is systematically higher than the experimental one in the visible range. This is to be expected when the film surface is not mirror-like : the reflectivity is more affected than the transmission, especially in the visible range.

	ε_∞	(cm^{-1}) ω_p	γ	ω_1	γ_1	$\omega_{\tau 1}$	ω_2	γ_2	$\omega_{\tau 2}$
S 1	3	8400	1300	450	150	4500	1000	7000	13900
S 2	3	9100	2100	580	300	3200	1000	7400	13700
	(cm^{-1})ω_3	γ_3	$\omega_{\tau 3}$	ω_4	γ_4	$\omega_{\tau 4}$	ω_5	γ_5	$\omega_{\tau 5}$
S 1	15500	7000	8600	21500	10800	15300	30000	9000	16000
S 2	15500	7000	9300	21500	11000	14600	30000	11000	18000
	(cm^{-3}) n		($\mu\Omega$.cm) ρ		(A) λ_1 (a,b)		(cm) ep1		ep2
S 1	7.73 10^{20}		1142		1895		3.6 10^{-5}		0.06
S 2	9.08 10^{20}		1572		1749		3. 10^{-5}		0.06

TABLE 1 Best fit parameters

ep1, ep2 are respectively the thickness of the thin film and of the substrate MgO

We have not been able to fit both transmission and reflectivity spectra without including the MIB (ω_2 = 1000 cm^{-1}, ω_{f2} = 13500–14000 cm^{-1}, γ_2 = 7000 cm^{-1}). Since the origin of this MIB is not known we consider it, in the present state of our knowledge, as a phenomenological description of the IR regime. The fit of the IR spectrum is then a balance between the Drude contribution and the MIB. This obviously decreases the accuracy of the parameters. Our best fit for the plasma frequency yields around 9000 cm^{-1}. The associated London penetration depth (within the CuO planes) is 1700 Å, assuming that all carriers participate to the superconducting properties.

We find a relaxation rate in the two samples which is definitely much larger than what has been reported in previous work : γ = 1300 and 2000 cm^{-1}. This result relies however on the assumption that the parameters of the MIB are the same in both films.

We observe that from the optics we find : ρ_1 = 1100 microhms and ρ_2 = 1500 microhm-cm, which tracks the 1150 and 1950 microhm-cm dc resistivities.

In the visible range, we identify three bands at 15300, 21500 and 30000 cm^{-1}. The most noticeable point is that transitions with very similar characteristic frequencies have been found in the $YBa_2Cu_3O_{6+x}$ compounds[12,13,14]. The existence of these transitions in the $Bi_2Sr_2Ca_1Cu_2O_8$ system as well strongly suggests that they share a common origin. More specifically, despite the variation of these bands with oxygen content x in $YBa_2Cu_3O_{6+x}$[15,16], our result could rule out the contribution of the Cu–O chains to these transitions.

6.CONCLUSION

By using simultaneously reflectivity and transmission data on two $Bi_2Sr_2Ca_1Cu_2O_8$ thin films, we have shown that in order to account for both spectra, the infrared response cannot be Drude-like, even at room temperature. The plasma frequency is then found to be 9000 cm^{-1}, which yields an estimate of the London penetration depth (1700 Å) associated to the Cu–O plane. Such room temperature spectra provide an easy and fairly accurate characterisation of the film resistivity. The simultaneous use of transmission and reflection data has allowed to show the existence in the $Bi_2Sr_2Ca_1Cu_2O_8$ compound of transitions very similar to the ones found in the $YBa_2Cu_3O_{6+x}$ system, suggesting that the origin of these transitions should be found in shared characteristic properties of these compounds.

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