Microscopic analysis of the transmittance of $YBa_2Cu_3O_7$ thin films

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The transmittance of a thin film of $YBa_2Cu_3O_7$ is calculated from the band-structure-derived dielectric function. It is shown that (1) band-structure calculations provide a reasonable description of the transmittance in the whole measured energy region ($0.2\ \text{eV} < \hbar\omega < 5\ \text{eV}$). (2) the transmittance in this region is mainly defined by the dielectric function corresponding to the light polarized in the x (crystallographic a) direction and (3) the minima of the transmittance do not necessarily indicate interband transitions.

Optical measurements on high-T_c superconductors (HTSC) provide valuable information about their electronic structure, but optical spectra are more difficult to interpret than many other spectroscopic techniques, like photoemission. In the optics of simple or transition metals the optimal way of dealing with the experimental spectra has turned out to be just the calculation of measured quantities from the band structure and the subsequent comparison with experiment (see, e.g., refs. [1,2]). Unfortunately, this method has not been used much in the case of high-T_c superconductors for two reasons: first, such calculations are much more complicated for HTSC, and, second and more importantly, the strong correlated nature of the electrons in these materials cast doubts on the relevance of band-structure calculations to optical measurements.

However, recent calculations have shown that traditional first-principles calculations are still able to describe reasonably most of the optical experiments in HTSC. One can mention, for instance, calculations for La_2CuO_4-based materials [3,4], for $YBa_2Cu_3O_6$ [5], and for $YBa_2Cu_3O_7$ [6]. Generally, for high-energy spectra ($\hbar\omega > 5$ eV) the agreement between experiment and theory is very good ([5,6]; see also ref. [7] for a comparison between the experiment and the calculations of ref. [3]). The agreement in the low energy region is more questionable. However, first principles calculations allowed one to explain qualitatively the peculiarities of the infrared optics of $(La,Sr)CuO_4$ superconductors, first of all the independence of the plasma frequency on doping. One also has to keep in mind that both the experiment *and* calculations are still improving, both being still *under* the standards used in the optics of usual metals [1,2].

Up to now, the comparison between calculations and measurements was done for reflectivity, for electron-energy-loss spectra or for ellipsometric spectra. There are however optical experiments on the transmittance of thin films [8,9] which are interesting in that they reflect features of the dielectric function in a manner different from the above-mentioned experiments. Experimentalists often plot the inverse transmittance $1/T$ [8] or its logarithm $\log(1/T)$ [9] in the hope that such plots correspond to the electronic absorption $\text{Im}\,\epsilon(\omega)$, which is not always true. In this paper we try to clarify the situation by calculating the transmittance of $YBa_2Cu_3O_7$, by analyzing it with comparison to the experiment.

The transmittance of thin metallic films is a complicated function of the dielectric function (DF). The full theory was given by Piro [10] for arbitrary anisotropy and angle of incidence, and both for smooth and rough films. Applying Piro's formulas to our calculated DF of $YBa_2Cu_3O_7$ shows that the transmit-

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tance (T) turns out to be nearly independent of parameters like the substrate dielectric constant or the angle of incidence (for $\theta_i < 15°$). As will be discussed below, for non-polarized light the transmittance is mostly defined by the x-component of the DF. Taking all this into account, one can rewrite Piro's formulas (which were actually used in our computer program) as

$$T = |\cos(\delta\sqrt{\epsilon_x}) - \tfrac{1}{2}i(\sqrt{\epsilon_x} + 1/\sqrt{\epsilon_x})\sin(\delta\sqrt{\epsilon_x})|^{-2}, \quad (1)$$

where $\delta = \omega d/c$, d is the thickness of the film, c is the velocity of light and $\epsilon_x(\omega)$ is the x-component of the complex DF. Eq. (1) is valid for smooth films. For rough films in the same limit we have in place of eq. (1)

$$T = \frac{(1-R)^2 \exp(-2\delta\sqrt{\epsilon_x})}{1 - R^2 \exp(-4\delta\sqrt{\epsilon_x})}. \quad (2)$$

Eqs. (1) and (2) coincide [#1] for δ (or d) $\to\infty$ and T is defined by the leading exponent:

$$T \propto \exp(-2\delta \operatorname{Im}\sqrt{\epsilon_x}). \quad (3)$$

Eq. (3) is useful for a qualitative analysis. First of all, let us look at T in the Drude limit where

$$\epsilon = \epsilon_D = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)}. \quad (4)$$

Direct calculation shows [#2] that the maximum of the expression $\omega \operatorname{Im}\sqrt{\epsilon_D}$, i.e. the minimum of T, occurs near $\omega = \gamma$. Taking into account that for HTSC γ is of the order of 1 eV, we come to the conclusion that the Drude behaviour is decisive in the region of interest (0.1 eV $\leqslant \hbar\omega \leqslant$ 4 eV). Indeed, comparing our calculations with and without interband contribution in the DF (fig. 1) we see that, generally speaking, the role of the interband transitions is to produce a small maximum near $\hbar\omega \sim 1$ eV and some structure at higher energies.

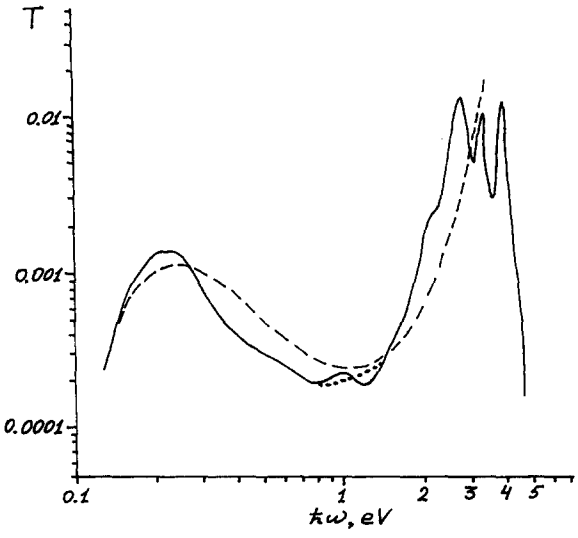

Fig. 1. Transmittance of the $YBa_2Cu_3O_7$ film for the light polarized in x-direction, $d = 2900$ Å, calculated Drude parameters (the plasma frequency was calculated by numerical integration of the squared Fermi velocity over the Fermi surface; the relaxation frequency γ was calculated from ω_p assuming the corresponding resistivity equal to 500 μΩ cm) are $\omega_p = 3.5$ eV, $\gamma = 0.83$ eV. The intraband (Drude) contribution is shown by the dashed line and the transmittance with the absorption maximum at $\hbar\omega = 1.1$ eV cut off (cf. fig. 3) by the dotted line.

Let us now turn to the experiment [8]. The authors of ref. [8] associated each minimum on the $T(\omega)$ curve with an electronic interband d–d absorption (0.6, 1.4 and 3 eV) or with charge-transfer excitations (4–5 eV). As is seen from fig. 2, the agreement between the calculated and the experimental positions of the minima is quite reasonable, the minima on the theoretical curve being situated at 0.8, 1.2, 3.2, 3.7 and decrease of T starting at 4.3 eV.

On the other hand, while the positions of the features on the experimental curve coincide with the theoretical ones, the magnitude of T is considerably lower in the latter case. One must keep in mind that the films used were not homogeneously thick and the thickness quoted in ref. [8] probably corresponds to the average thickness while T is mainly defined by the minimal thickness. Then, the measured T varies extremely weakly, taking into account the exponen-

[#1] However, for $d \sim 3000$ Å the difference still exists. We used actually formulas for smooth films.

[#2] The underlying band structure was calculated by the LMTO method with combined corrections [11]. On the Y, Ba and Cu sites angular momenta up to f were included and on the O and empty-sphere sites up to d. The optical matrix elements were calculated as described in ref. [2] but without combined correction to the matrix elements themselves.

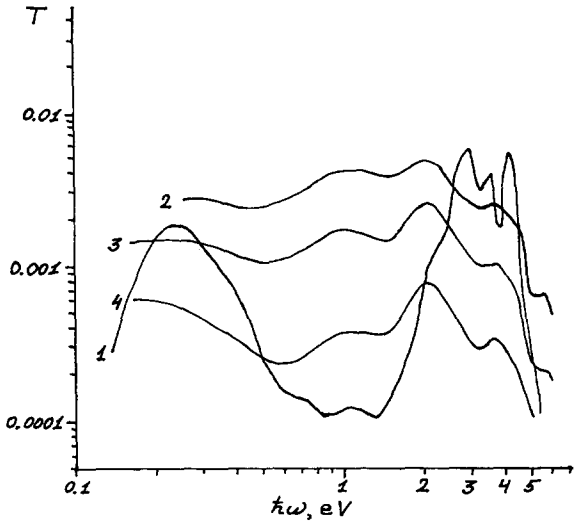

Fig. 2. Experimental [8] and calculated (averaged for x- and y-polarizations) transmittance of the $YBa_2Cu_3O_7$ film: thickness $d=2900$ Å in calculations (1) and $d=2900$ Å (2); $d=3400$ Å (3); $d=4300$ Å (4) in the experiment. Drude parameters for the y-polarization are $\omega_p=4.2$ eV, $\gamma=1.2$ eV.

tial dependence of T on $\epsilon(\omega)$. We do not have an explanation of this fact but believe that this is an *artefact* of the experiment because, as will be discussed below, any reasonable Drude parameter set would already without any interband part produce T varying by not less than an order of magnitude. Also if we just use in (1) or (2) the dielectric function derived from reflectivity or ellipsometry experiments we would have much stronger variation of T. Anyway, one can be much more sure in the position of the minima of T, so later we shall discuss the experiment only from this point of view.

The good agreement between the theory and the experiment for $\hbar\omega > 3$ eV corresponds to earlier findings [5–7] that the band-structure calculations provide a good description of the optics of HTSC in this energy region; the analysis of the optical absorption for $\hbar\omega > 3$ eV is given elsewhere [6]. Here we shall concentrate on the lower energy region which is hardly possible to study by traditional ellipsometry and difficult to interpret in the reflectivity measurements. At the same time, as we shall see, the transmittance measurements serve as a "microscope" which allows one to look at the region near $\hbar\omega \sim \gamma$.

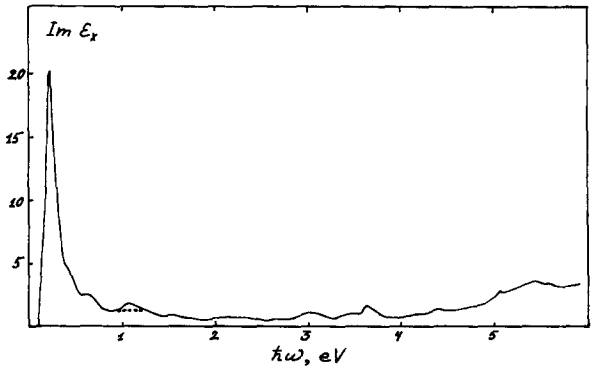

Fig. 3. Imaginary part of the interband dielectric function $\mathrm{Im}\,\epsilon_x(\omega)$. The dotted line shows the same quantity with the maximum at 1.1 eV cut off.

If we look at the curve $\mathrm{Im}\,\epsilon_x(\omega)$ (fig. 3) in this region we see that the only feature here is a relatively weak absorption maximum at $\hbar\omega = 1.1$ eV ($\mathrm{Im}\,\epsilon_y(\omega)$ has different structure but it is much larger than $\mathrm{Im}\,\epsilon_x(\omega)$ and hence does not manifest itself in T). Indeed, cutting off this maximum (fig. 3) results in the disappearance of the minimum in T at $\hbar\omega \sim 1.2$ eV and therefore in the disappearance of any qualitative difference between the total and the intraband T in the region $\hbar\omega < 3$ eV [#3]. The minimum at $\hbar\omega \approx 0.8$ eV is therefore not due to some interband transition but arises just because of the interference between the intraband and the interband T. In other words, the analysis above and comparison to the intraband-only T (fig. 1) shows that the positions of minima of T not always have a direct physical meaning.

To conclude, we calculated the transmittance of $YBa_2Cu_3O_7$ films, using the band-structure derived DF [6]. The result confirms our earlier conclusion [6] that the latter provides a good description of the optical properties at $\hbar\omega > 3$ eV, and proves the applicability of such an approach also for smaller energies. We also showed that the minima on the curve $T(\omega)$ do not necessarily correspond to the interband absorption maxima, i.e. to the electronic transition, but may appear as a result of an interplay between

[#3] It is hard to imagine that such a weak feature on the background of a large (at this energy) intraband absorption can be seen in any other optical experiment.

the interband and intraband absorption.

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