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Unexpected behaviour of IR reflectivity of a $YBa_2Cu_3O_{7-\delta}$ oriented film

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Abstract

a-b plane oriented $YBa_2Cu_3O_{7-\delta}$ films have been obtained by laser ablation. Electrical and structural characterization is presented as well as temperature dependent (4 K up to room temperature) optical reflectance spectra recorded between 50 cm^{-1} and 22 000 cm^{-1}. Extra absorptions at low frequencies and low temperatures are analysed and their connection with a possible superconducting gap is discussed. It is emphasized that the overall conducting behaviour in the normal state is well described by a single Drude-like modified term.

Keywords: YBCO thin films, Optical conductivity, Superconducting gap

1. Introduction

Since the discovery of high temperature superconductivity in layered copper-based oxides by Bednorz and Muller [1], an enormous effort to understand the physics of these compounds has been made. An even greater interest in this subject has come with the synthesis by $YBa_2Cu_3O_7$ (YBCO), having a critical temperature above liquid nitrogen temperature [2]. One of the main features of layered oxide superconductors is the highly anisotropic, even bidimensional, conducting behaviour: the charge carriers are mainly confined within the basal *a*-b plane [3]. Thus, the obtention of good quality YBCO *a-b* oriented films, independent of their interest for future applications, is also of great interest for the understanding of their conducting properties.

A very powerful tool to study these systems is the IR (and more generally optical) reflectivity. IR-visible spectroscopy enables us to obtain information about, in addition to phonons, charge carriers (plasmons), superconducting gap and higher energy (mid-IR) absorptions. Although the phonon behaviour is nowadays well understood in YBCO [4,5], the other features observable in the IR and visible spectrum of this material are far from giving rise to any consensus yet. (i) The plasmon in YBCO cannot be fitted with a simple Drude expression [6 12]. (ii) The explanations of mid-IR excitations are unclear [3, 7, 8]. (iii) The existence of an observable superconducting gap in the Bardeen-Cooper-Schrieffer (BCS) sense is controversial [6, 8, 19].

In this work, we present optical reflection over a YBCO film grown by laser ablation, between 4 K and room temperature. Spectra have been recorded with a single spectrometer covering the spectral range from 50 to 22 000 cm^{-1}, thus including all the excitations discussed above.

2. Experiment

The spectra have been recorded with a Fourier transform IFS 307 Bruker spectrometer composed of two interferometers with complementary spectral ranges, which cover the region from 8 cm^{-1} to 40000 cm^{-1}. Reference measurements have been done over an aluminium mirror and the reflectance error in our spectra does not exceed 2%. Temperature control accuracy, with the aid of a CLTS probe, was of the order of 0.5 K. The YBCO film has been deposited by pulsed laser ablation. The laser source is an excimer laser operating at 248 nm (KrF). After spatial filtering, the laser beam impinges on a bulk superconducting target. The vaporized material ejected from the target surface is deposited on a (100)-oriented single crystal of MgO substrate located opposite to the target. The film has a surface area of about 1 cm^2 and is 325 nm thick. Its X-ray diffraction pattern reproduced in Fig. 1 shows that it is highly *a-b* plane oriented (better than 0.7 °) and allows the determination of *c* lattice parameter: 11.695 ± 0.005 Å. The electrical resistivity has been measured by a standard DC four-probe technique. Thermal evolution of the DC resistivity is shown in Fig. 2. The superconducting transition has a critical temperature T_c = 90.5 K and a transition width ΔT = 0.6 K. A linear increase in resistivity is observed from T_c up to room temperature. The critical current density reaches a value as high as $5 \times 10^6 A.cm^{-2}$. All these data indicate a "δ" of about 0.05 and a relatively good quality film.

3. IR data fitting

Previous studies [6-10] have shown that a Drude term alone cannot explain the conducting behaviour of the *a-b* plane conductivity in YBCO. The best compromise with just a Drude term for the dielectric susceptibility $\chi_{plasmon}$ obtained in terms of the plasma frequency Ω_p and its damping γ_p, i.e.

$$\chi_{plasmon} = -\varepsilon_\infty \frac{\Omega_p^2}{\omega(\omega - i\gamma_p)} \qquad (1)$$

is shown in Fig. 3 and clearly is unable to fit the experimental result. Here Ω_p is defined in such a manner that, in the absence of damping, one obtains unity reflection up to $\omega = \Omega_p$. Note that in some reports ε_∞ is included in Ω_p^2. In the best compromise obtained with this model, the input parameters were Ω_p = 9100 cm^{-1}, γ_p =2500 cm^{-1} and ε_∞ =3.60.

Many models have attempted to reproduce the spectra with various phenomenological or microscopic bases. (i) The only model that keeps the Drude term requires several additional strong oscillators in the phonon and also mid-IR regions [6, 8, 11]. However, the dielectric strength of certain of these oscillators is unrealistically high for phonons (below 700 cm^{-1}), compared with phonons in O_6 compounds, for instance, and their origin is quite unclear in the mid-IR. In addition, the Drude contribution is not dominant. This seems physically artificial because, in a highly conducting medium, all oscillators are expected to be screened by charge carriers. (ii) Another way to fit the response in YBCO is to consider a frequency dependence of at least one term, either the scattering rate or the effective mass [5, 7, 20]. (iii) It is also possible to explain the plasmon-like response in YBCO as multiple contribution of different Drude terms but here again the microscopic origins are unclear [9, 10].

Here we use an extension of the Drude model, which introduces a very simple frequency dependence in the plasmon scattering rate. This model has already been successfully used to fit IR spectra of other conducting oxides such as reduced TiO_2 [21, 22], NbO_2 [23] and strontium titanate [24]. According to the Maxwell equations, transverse optical (TO) and longitudinal optical (LO) modes are the poles and zeroes respectively of the dielectric response. Hence the dielectric function model consists in factorizing over TO and LO modes [21]. The basic point added by such an extension is the possibility of having different damping terms of TO and LO modes. This possibility of different scattering rates has been shown to be very useful in cases where we have wide reflection bands (a large LO - TO splitting) [21-25]. If we regard the charge carrier contribution to the dielectric function as an additional "oscillator" with it's TO frequency equal to zero (no restoring force constant for mobile charge carriers) and the plasma frequency as an LO mode, we obtain an extension of the Drude contribution for the dielectric susceptibility $\chi_{plasmon}$ in the form

$$\chi_{plasmon} = -\varepsilon_\infty \frac{\Omega_p^2 + i(\gamma_p - \gamma_0)\omega}{\omega(\omega - i\gamma_0)} \qquad (1)$$

In Eq. (2), Ω_p is the plasma frequency as defined in Eq. (1) and γ_p and γ_0 are the dampings evaluated at Ω_p and in the static limit respectively. Setting $\gamma_p = \gamma_0$ transforms Eq. (2) into the commonly used Drude expression Eq. (1). Eq. (2) implicitly introduces a monotonic variation in the plasmon damping function from γ_0 at zero frequency to γ_p at the LO plasma frequency, whereas the usual Drude term Eq. (1) assumes a constant damping. More details about this model were recently given in Ref. [24].

3.1. The room temperature normal state

Fig. 3 shows spectra obtained at room temperature and 4 K. Above ca. 700 cm^{-1}. The difference between spectra is negligible within experimental error. This is unexpected because Fig. 2 shows a (common) temperature dependence of the DC conductivity and, therefore, the absence of significant dependence in the IR raises a problem. The overall behaviour of the room temperature spectrum can be obtained from the simple modified Drude term Eq. (2) with no additional feature. Phonons are completely screened by the plasmon, which is expected in view of the T_c of our sample, and so there is no need to introduce any phonon oscillator. (In fact, their introduction, with parameters known for the O_6 compound, does not significantly modify the response). The modified-Drude fit shown in Fig. 3 has Ω_p =9100 cm^{-1}, γ_p =7350 cm^{-1}, γ_0 =1750 cm^{-1} and ε_∞ = 3.60, values comparable with those found by Thomas et al. However, in Ref. [6], Thomas et al. used an additional mid-IR oscillator. The high damping values (when compared with Ω_p), which are observed in virtually all conducting oxides, may be regarded as a strong interaction between change carriers and crystalline lattice. We are, then, allowed to think that conduction mechanisms in these systems are polaronic (trapped rather than free charge carriers).

We are tempted, here, to make a parallel between the present model and other models dealing with the same problem. The marginal Fermi liquid (MFL) [26], the nested Fermi liquid (NFL) [27] and the antiferromagnetic Fermi liquid (AFL) [28] models are the most important examples. The MFL model is based on the hypothesis that there are excitations, which contribute to both spin and charge polarizability. The form of this excitation is stated to behave as ω/T when $|\hbar\omega| < k_BT$ and to be constant when $|\hbar\omega| > k_BT$. The IR data are then fitted by means of a Drude-like expression having frequency-dependent damping and frequency-dependent charge carrier effective mass. The NFL model, as its name states, requires the nesting of the Fermi surface and is also based on a Drude-like expression for optical conductivity. Both models predict a linear frequency dependence of damping. Both models fit quite well the frequency-dependent conductivity and are compatible with the linear thermal dependence of DC resistivity. The AFL Model is based on antiferromagnetic spin fluctuation interactions. If the model is less successful in describing IR-visible conductivity, it is still compatible with the thermal dependence of DC conductivity.

The model used in this paper also describes in a quite accurate manner the frequency dependence of optical conductivity. It is not possible to compare its formalism directly with the other models. The only situation in which we can do so is when $\gamma_p - \gamma_0 << \Omega_p$. In this approximation one obtains $\gamma(\omega) \approx \gamma_0 + (\gamma_p - \gamma_0)\ \omega^2/\omega_p$ for the present model. The quadratic term is then very weak and may be considered as a slight perturbation in the Drude classical form. However, when $\gamma_p - \gamma_0 \approx \Omega_p$ (as is the case in the present work) the functional form of IR conductivity can no longer be compared with the Drude Model. The present model has already proved itself able to fit data from various conducting systems [21-25] and we must emphasize that in YBCO, contrary to some analyses of other experimental data [6, 8, 29], the overall response is here obtained via this single modified Drude-like term Eq. (2). This YBCO film needs no additional contribution in the phonon or mid-IR regions. This means that, if such mid-IR absorptions appear at lower electronic concentration (lower oxygen content), they are masked here by the plasmon as well as phonons are screened. Moreover, no assumption about particle interactions (magnetism, low frequency dependence) or system configuration (nesting of Fermi surface) is made, making the present model quite general. If the other models describe optical data in YBCO with the same accuracy, applying them to other systems (especially non-cuprate systems) can lead to some physical inconsistency. The present model, on the contrary, can be used in any conducting system.

3.2. The superconducting state

At low frequency, the liquid helium temperature spectrum exhibits an additional reflection band which is absent at room temperature. The thermal evolution of the low frequency spectrum of our film is displayed in Fig. 4. Fig. 4 also shows a comparison of the reflection at all temperatures with the 300 K reflectivity at three different frequencies within the 200 400 cm^{-1} range. Below 200 cm^{-1}, experimental uncertainties limit the accuracy of the data. Above 400 cm^{-1}, the change in spectral data is too small with respect to the experimental error. Looking higher in frequency, we see that the spectrum at 4 K follows the normal state spectrum down to about 600 cm^{-1} and then has an enhanced reflectivity for the lower frequencies. The problem, thus, is to determine whether this departure starts at the superconducting transition temperature or not. In other words, we want to see whether this behaviour can be related to the condensation of Cooper pairs or whether it is just continuous evolution of the spectra. The plot of $\Delta R/R_{300}$ in Fig. 4 seems to be clear about the behaviour, at least at intermediate frequencies. Between 300 K and 102 K, no enhancement of the reflectivity is observed. The reflectivity for temperatures between 4 K and 90 K also is clearly higher than the normal state reflectance. Within experimental uncertainty and on the basis of this plot in Fig. 4, it appears that this change to regime starts at the transition temperature. We can conclude, then, that this reflection band is related to the superconducting transition and thus to the formation of Cooper pairs. In BCS theory, the formation of a superconducting gap is expected. The IR response of such a gap is a flat unity level in reflectivity below the gap energy and a thermal evolution of this energy gap (the gap being the order parameter, we should expect a second-order transition behaviour). This is not what we observe in our sample. No flat unity level has been attained and no frequency shift of our onset frequency has been observed. Anyway, one may consider that a standard BCS gap is unlikely to exist in YBCO as, at least, phonon states lie within this "gap" region. We will not enter in a discussion of *s* wave or *d* wave superconductivity but, if we insist (for comparison with other data) on taking a frequency (about 500 cm^{-1}) just below this onset in terms of a BCS-like gap, we find $2\Delta / k_B T_c = 8$, as already reported [30, 31]. This value is also consistent with indications deduced from some other methods (nuclear magnetic resonance, Raman photoemission) [32-36]. Present experiment data are very similar to those reported by Kamarfis et al. [8] below 100 K, but contrary to their report, we do not observe any significant evolution of the reflectivity between 100 K and room temperature. They did not relate this low frequency increasing in reflection to a superconducting gap as they considered that a residue of this profile was still present at room temperature. This is not the case in our film. The extra band is clearly related to the superconducting transition

3.3. Optical conductivity

Fig. 5 shows the real part of the optical conductivity of our sample. The theoretical curve for room temperature spectrum has been obtained from Eq. (2) and the curves at 4 K and 300 K are Kramers Kronig transformations of reflection data in the superconducting and normal states respectively. Adopting different extrapolations to zero frequency leads to slightly different low frequency behaviours in conductivity. To avoid misinterpretation of data we present Kramers Kronig transformations above the 380 cm^{-1} region which, in our case, was independent of the low frequency extrapolation. Once again, the agreement between the model and the experiment is quite satisfactory. Moreover, the extrapolation of the fitted conductivity to zero frequency falls into the same range of DC conductivity measured for similar films [37]. In the experimental spectra, one sees the effect of the superconducting transition in the frequency region below 600 cm^{-1}while in the normal state the conductivity raises continuously on decreasing frequency, in the superconducting phase it drops for frequencies below 600 cm^{-1}, an effect possibly linked to the superconducting transition. This possible relation is even more evident in Fig. 6 where we present conductivity obtained from Kramers Kronig transforms. No significant change has been observed between 300 K and 102 K. On the contrary, a sudden depart from this "room temperature behaviour" can be inferred below T_c for low frequencies.

4. Conclusion

In this work, we have presented the characterization of a $YBa_2Cu_3O_7$ film obtained by laser ablation. The aim of high critical current densities has been achieved in this film, which presents an almost single-crystal structure configuration with a high *a-b* plane orientation. We have characterized the temperature dependence of the reflectivity and focused the low frequency extra absorptions that develop below T_c. Present results and their analysis follow a long-standing controversy about both of the main discussed points (superconducting gap and charge carrier response). We show the onset of an additional band that appears very probably at T_c and increase below. From both reflectivity and conductivity curves, we can associate this change in reflectivity with a superconducting condensate. If the cut-off frequency of this feature is assigned to a superconducting gap, then one confirms results previously obtained by others and commonly interpreted in terms of $2\Delta / k_BT_c = 8$. We have also been to fit the normal state spectrum with a single modified Drude-like term (no additional phonon or mid-IR contributions) which assumes a frequency-dependent damping or, equivalently, a frequency-dependent carrier effective mass.

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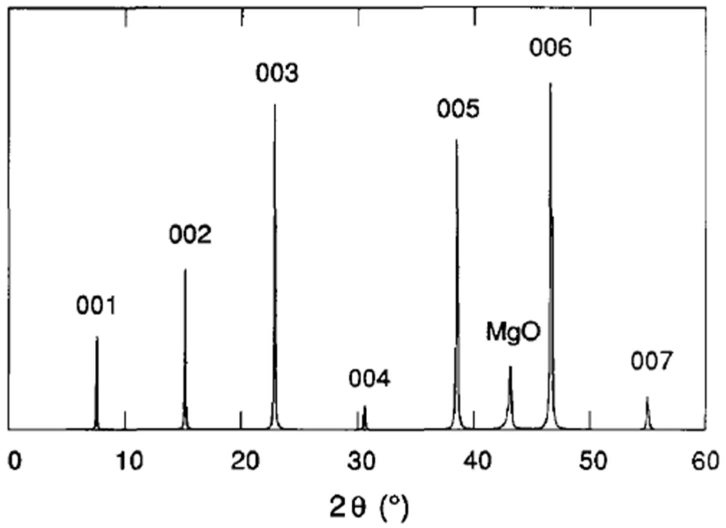

Fig. 1. X-ray diffraction pattern of YBCO film showing its (001) orientation.

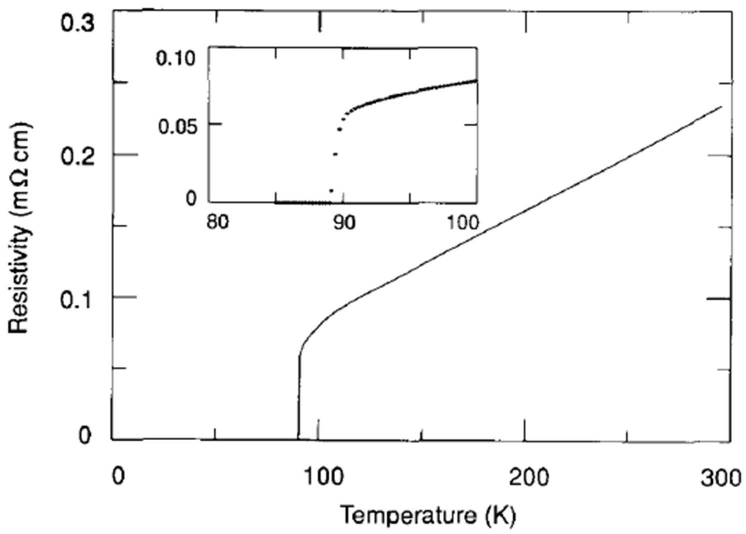

Fig. 2. DC resistivity of YBCO film. The 0.6 K width of the superconducting transition is also shown.

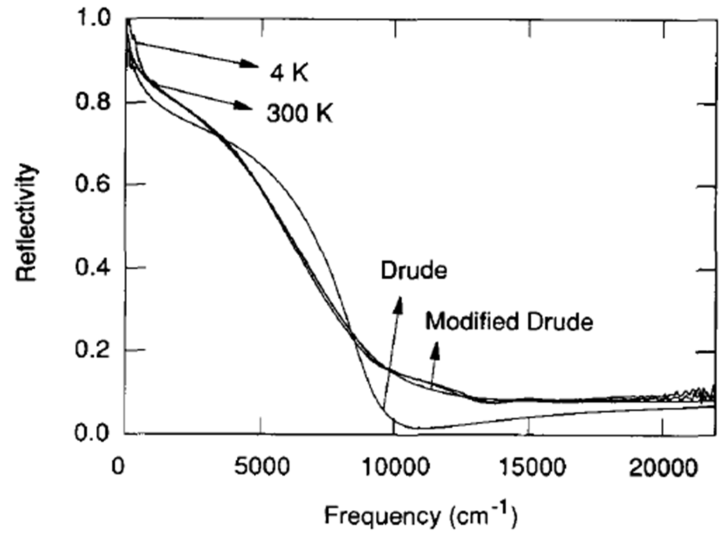

Fig. 3. IR and visible reflection of YBCO film at 4 K and 300 K and best fits obtained from a simple and a modified Drude behaviour. Fit parameters are given in the text.

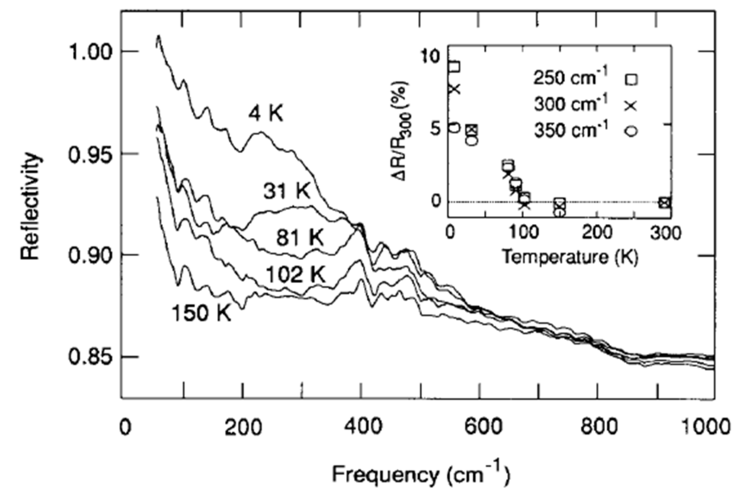

Fig. 4. Low frequency behaviour of IR reflection in YBCO film. A comparison is also shown of the reflectivity at temperature T with the room temperature value via [R(T)- R(300 K)] / R(300 K) for three frequencies in the low energy part of the spectra. The uptake in reflectance below 600 cm^{-1} can be associated with the superconducting transition.

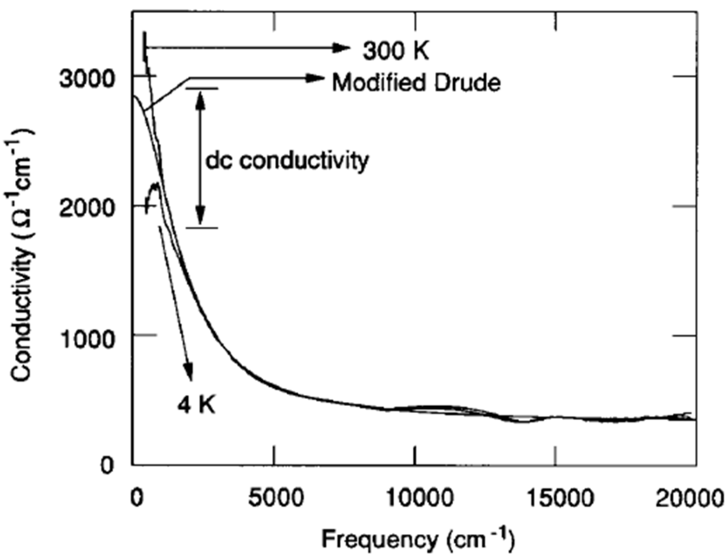

Fig. 5. Real part of IR and visible conductivity. Data at 4 K and 300 K have been obtained from Karmers Kronig transforms of reflection data and theoretical curve is obtained from our modified Drude expression with the same parameters used in Fig. 2. Double arrow shows DC conductivity measured by Flik et al. [37] for a set of similar thickness range films.

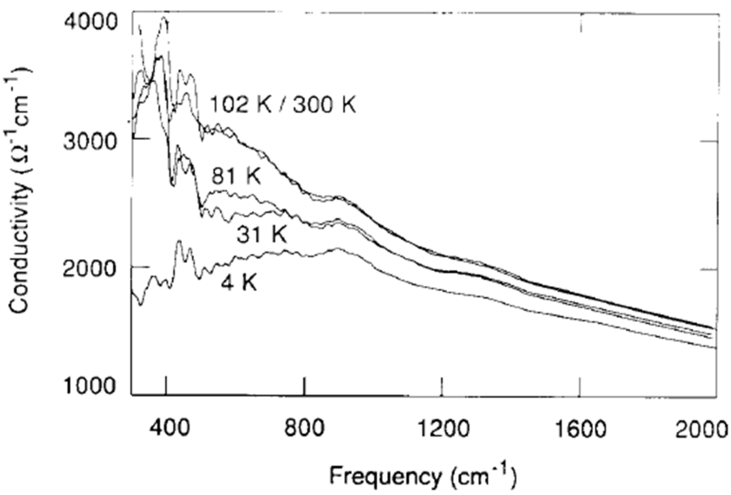

Fig. 6. Low frequency dependence of real part of conductivity between 4 K and 300 K. Departure from a "'room temperature behaviour" occurs at T_c and can be related to the superconducting transition.