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INFRARED STUDIES OF AB-PLANE ORIENTED $YBa_2Cu_3O_{7-\delta}$

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Measurements of the infrared properties parallel to the two-dimensional conducting Cu-O layers of the high-Tc compound $YBa_2Cu_3O_{7-\delta}$have been carried out between 300 K and 4 K. Samples have included textured ceramics, oriented films, and small crystals. A strong, temperature-independent absorption is observed in the mid-infrared, with a temperature-dependent Drude contribution in the far infrared. In the superconducting state most of the oscillator strength of the Drude absorption moves to zero frequency and the reflectance spectrum is dominated by a plasma edge at 500 cm^{-1}. A superconducting gap cannot be seen in our spectra.

I. Introduction

There has recently been considerable attention to the infrared properties of the high-temperature superconductors[1–12], focussing on both the superconducting energy gap and question of the presence of a strong non-Drude absorption in the mid- infrared. In this paper we present the results of a detailed study of the the infrared properties over a wide range of frequencies and temperatures in ab-plane oriented samples of $YBa_2Cu_3O_{7-\delta}$. We find that the mid-infrared region is dominated by a strong, nearly temperature-independent absorption band, possessing around 80 % of the infrared oscillator strength. In addition, at temperatures above the superconducting transition temperature, a Drude contribution is observed in the far infrared; the temperature dependence of this Drude term is in agreement with the dc conductivity.

Below T_c the Drude contribution has disappeared, suggesting that most of the carriers responsible for the dc transport have condensed to form a collective state. A finite residual absorption remains at low temperatures, possibly the low frequency tail of the mid-infrared band; and no energy-gap structure can be seen in the frequency dependent conductivity of our samples. Although a peak is seen at 500 cm^{-1} in the ratio R_s/R_n of superconducting to normal reflectance, this peak is not the energy gap, but instead can be associated with a plasmon-like mode of the superconducting electrons, sufficiently screened by the positive contribution to the dielectric function of the mid-infrared absorption band that it appears in the far infrared rather than at the bulk plasma frequency.[1,2]

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II. Experimental details

Our measurements have been made on individual (twinned) crystals, on mosaics of small crystals, on oriented films, and on "textured"[3] ceramic samples. The crystal mosaic consisted of ten to twenty small flux grown crystals, each 300 to 1500 μm in size. On account of the well-known ab twinning which occurs in $YBa_2Cu_3O_{7-\delta}$crystals, the reflectance of each crystal in the mosaic is an average of the a-axis and b-axis response. The films were grown on (001) $SrTiO_3$ substrates and had transition temperatures near 87 K. The textured ceramics were as-grown surfaces of pressed-pellet samples having a high degree of orientation, with about 80 % of the surface consisting of tile-like crystallites, 10 μm in size, with their a and b axes lying in the surface.

We measured the the far-infrared through near ultraviolet reflectance of our samples between liquid helium and room temperature. Following the measurement, the surfaces of the sample were coated with an ordinary metal (Pb or Al) and the reflectance of the coated surface was measured, in order to correct for imperfect sample surfaces. Because of the the incomplete parallelism of the crystals making up the mosaic and rough surfaces of the textured ceramics this coating was an extremely important part of the measurement procedure. The frequency-dependent conductivity and dielectric function were determined by a Kramers-Kronig analysis or by least squares fits to model dielectric functions.

III. Results

We find two principal contributions to the ab-plane infrared conductivity of $YBa_2Cu_3O_{7-\delta}$: a Drude part in the far infrared and a mid-infrared band. In addition, in films and in textured ceramics, there is an additional contribution of sharp lines from phonons. Weak phonon structure is also seen in the crystal data.

These are illustrated in Fig. 1, which shows the 300 K frequency-dependent conductivity of a thin film. Our reflectance data are similar to those of Ref. 8. Three difficulties are apparent in attempting to interpret these data within a single Drude model, for which

$$\sigma_1(\omega) = \frac{\sigma_{dc}}{1+\omega^2\tau^2} \tag{1}$$

where σ_{dc} is the ordinary dc conductivity and τ the electronic relaxation rate. First, $\sigma_1(\omega)$ exhibits a broad mid-infrared peak spanning roughly 0.1 to 1 eV, differing from the monotonic decreasing $\sigma_1(\omega)$ of the Drude model. Second, the values of τ required to force-fit the data give unrealistic scattering rates, as discussed below. Finally, the mid-infrared $\sigma_1(\omega)$ is essentially temperature independent in disagreement with the strongly temperature-dependent dc conductivity, for which $\sigma_{dc} \sim T^{-1}$.

To illustrate the various contributions to $\sigma_1(\omega)$, we show in Fig. 2 the data for a textured ceramic sample. The smooth curve is the conductivity calculated from a fit to the measured reflectance of a dielectric function of the form:

$$\epsilon_n(\omega) = -\frac{\omega_{pD}^2}{\omega^2 + i\omega/\tau} + \sum_{j=1,6} \frac{S_j\omega_j^2}{\omega_j^2 - \omega^2 - i\omega\gamma_j} + \frac{\omega_{pe}^2}{\omega_e^2 - \omega^2 - i\omega\gamma_e} + \epsilon_\infty \tag{2}$$

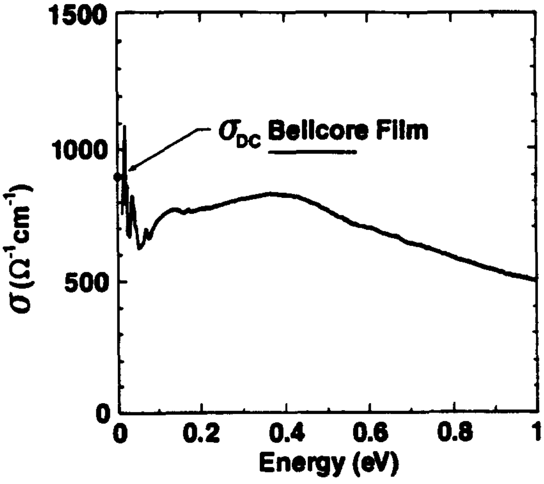

Fig. 1. The frequency-dependent conductivity at 300 K of a thin film of $YBa_2Cu_3O_{7-\delta}$ obtained by Kramers-Kronig transformation of the reflectance.

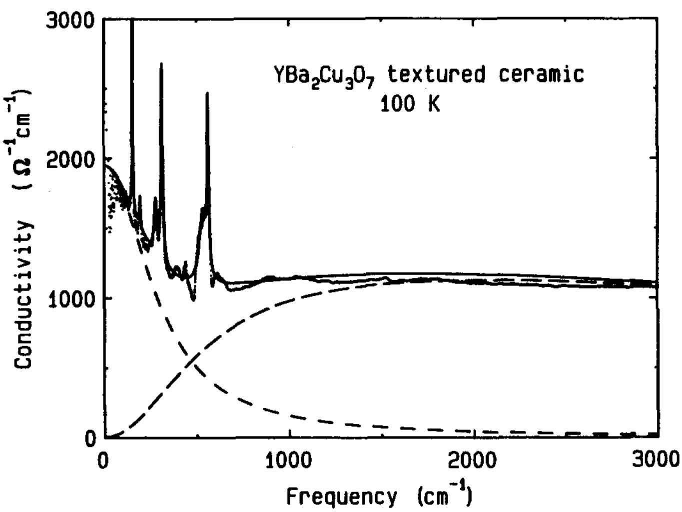

Fig. 2. The 100 K conductivity of a textured ceramic sample of $YBa_2Cu_3O_{7-\delta}$ obtained by Kramers-Kronig transformation of the reflectance is shown as points. The smooth curve is a fit of the reflectance to the dielectric function of Eq. 2. The dashed curves shows individual contribution of the Drude and the mid-infrared bands to the conductivity.

where the first term is a Drude contribution characterized by plasma frequency $\omega_{pD} = 5900\ cm^{-1}$ (0.74 eV) and relaxation rate $1/\tau = 300\ cm^{-1}$ (0.037 eV); the second is from six optical phonons, with frequencies ω_j, damping constants γ_j, and oscillator strengths S_j; the third the contribution of the mid-infrared absorption band, represented by a highly overdamped oscillator with center frequency $\omega_e = 2100\ cm^{-1}$ (0.26 eV), strength $\omega_{pe} = 21{,}000\ cm^{-1}$ (2.6 eV), and width $\gamma_e = 8400\ cm^{-1}$ (1 eV); the last the high-frequency value of ϵ_1, here taken to be $\epsilon_\infty = 3.8$. The individual contributions of the far-infrared Drude term and the mid-infrared absorption are shown as dashed lines in Fig. 2.

The far-infrared Drude conductivity is consistent with the dc conductivity, both in magnitude and in temperature dependence. In fact, the resistivity ρ measured by far-infrared means (the inverse of the $100\ cm^{-1}$ conductivity) is linear in temperature, with a relatively small zero temperature intercept, typical of high-quality $YBa_2Cu_3O_{7-\delta}$.

Fig. 3 shows the normal- (100 K) and superconducting-state (10 K) reflectances for a single crystal mosaic. A simple model fit to the 100 K data is also shown: a Drude absorption at zero frequency with $\omega_p = 9700\ cm^{-1}$ (1.2 eV) and $1/\tau = 250\ cm^{-1}$ (0.03 eV) and a mid-infrared band represented as before with a single broad oscillator. This oscillator has center frequency $\omega_e = 1500\ cm^{-1}$ (0.19 eV), strength $\omega_{pe} = 20{,}700\ cm^{-1}$ (2.6 eV), and width $\gamma_e = 3600\ cm^{-1}$ (0.45 eV). Calculations of reflectance which use only a Drude term are also shown in Fig. 3. The higher reflectance curve uses parameters that fit the far infrared (given above) while the lower curve fits the mid-infrared region with a single Drude expression[7,8] with $\omega_p = 25{,}000\ cm^{-1}$ (3.1 eV) and $1/\tau = 4100\ cm^{-1}$ (0.5 eV). It is clear that a single Drude expression cannot fit the whole curve.

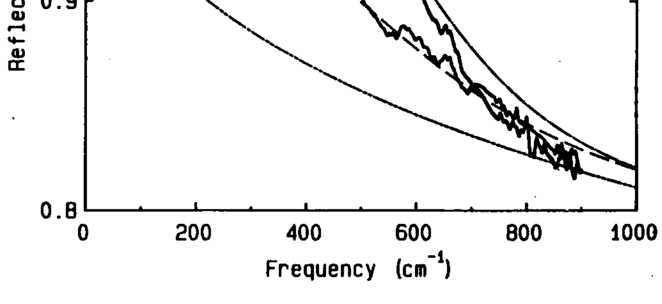

Fig. 3. Reflectance of a single crystal mosaic of $YBa_2Cu_3O_{7-\delta}$ at 100 K in the normal state and at 2 K in the superconducting state (solid, thick curves). The dashed curves show various model fits described in the text: — — — Drude plus mid-infrared oscillator for the normal state, — - - — - - — Drude fit to mid infrared, - - - Drude fit to far infrared, — - — - — plasma model for the superconducting state.

IV. Discussion

A. Normal-state properties.

The fit to the reflectance of the crystals shows that 80 % of the infrared oscillator strength is in the mid-infrared band, with only about 20 % in the low frequency Drude absorption. The total oscillator strength, given by $(\omega_{pD}^2 + \omega_{pe}^2) = (2.9 \text{ eV})^2$, is close to what is found by earlier fits to a single Drude model, which found[7,8] ω_p = 25,000 cm^{-1} (3.1 eV) and $1/\tau$ = 7500 cm^{-1} (0.9 eV).

We can estimate the mean free path from the relaxation time found in the fit. Using $\ell = v_F\tau$ and taking $v_F = 2\text{x}10^7$ cm/sec, our relaxation rate of $1/\tau$ = 300 cm^{-1} yields ℓ = 35 Å. This is a much more reasonable value than the 1 to 2 Å mean free path estimated from the $1/\tau$ = 4100 cm^{-1} required to fit the reflectance to a single Drude dielectric function.

B. Superconducting state properties.

Because an ordinary superconductor has no loss for photon energies smaller than the energy gap, 2Δ, the reflectance is unity below the gap, falling to join the (Drude) normal-state reflectance at frequencies substantially above the gap. The ratio R_s/R_n has a maximum near 2Δ. Attempts to use measurements of R_s/R_n to estimate the superconducting gap in the new high-T_c materials by analogy to ordinary superconductors have been made by many groups, with estimates of $2\Delta/k_BT_c$ ranging from 1.5 to 8. However, the high T_c materials are not ordinary metals; the dielectric function has complicated behavior. In superconducting $La_{2-x}Sr_xCuO_4$a plasma edge develops in the far infrared, which looks deceptively like a BCS gap edge.[1,2] However, these changes in reflectance can be traced to the real part of the dielectric function $\epsilon_1(\omega)$, which is large and negative at very low frequencies (due to the zero frequency delta-function response of the superfluid electrons) but which becomes nearly zero in the far infrared (due to screening by the large positive contribution to $\epsilon_1(\omega)$ from phonons and the mid-infrared band.)

Following Sherwin et al.,[1] we use a simple zero-parameter model, the plasmon model, to describe the superconducting state in $YBa_2Cu_3O_{7-\delta}$. This model assumes that all of the oscillator strength which in the normal state resided in the Drude band of width $1/\tau$ has been shifted to a delta function at zero frequency. The model does not involve any particular value for the gap so long as $2\Delta \gg 1/\tau$. In addition, for simplicity the phonon terms have not been included in the dielectric function for the superconductor, $\epsilon_s(\omega)$, given by:

$$\epsilon_s(\omega) = -\frac{\omega_{ps}^2}{\omega^2} + i\frac{\pi\omega_{ps}^2}{2\omega}\delta(0) + \frac{\omega_{pe}^2}{\omega_e^2 - \omega^2 - i\omega\gamma_e} + \epsilon_\infty \qquad (3)$$

Here, ω_{ps} is the plasma frequency or oscillator strength of the superfluid electrons $\omega_{ps} = 4\pi n_s e^2/m$ with n_s the superfluid density. The second term in Eq. (3) is the delta function dc conductivity of the superconductor. The dash-dot line in Fig. 3 shows R_s calculated from the plasmon model. Note that the reflectance fall off beginning around 500 cm^{-1} is caused by the real part of ϵ_s approaching zero rather than by any onset of absorption.

Fig. 4 shows the ratio R_s/R_n of the crystal mosaic. The shape of the data is in good agreement with other results[7] on ab-plane crystal surfaces. A band of 2 – 4 % strength can be seen peaking at 500 cm^{-1}. The plasmon model calculation is also shown. The model describes the position of the peak correctly but it is important to stress that because there remains a low frequency absorption in our sample, the reflectance does not reach the expected 100 % at low frequency.

The changes seen in the reflectance on passing through the superconducting transition are due to the transfer of most of the oscillator strength of the Drude-like normal electrons to the zero-frequency delta-function response of the condensate. In the clean limit, the width of the Drude $\sigma_1(\omega)$ is smaller than the gap, making the magnitude of the conductivity at $\omega = 2\Delta$ small. In addition, there remains a finite conductivity from the mid-infrared absorption to further diminish the importance of any contribution from the superconducting electrons at and above the gap. Thus there is no sharp feature in the clean-limit reflectance at the gap frequencies; the far-infrared reflectance is determined by the inductive response of the condensate and the reactive and absorptive contribution of the mid-infrared band.

It should be emphasized that the plasma edge seen in the superconducting state at 500 cm^{-1} is a direct consequence of the dispersion caused by the strong mid-infrared oscillator strength. Other data on $YBa_2Cu_3O_{7-\delta}$crystals R_s/R_n data show a maximum near 500 cm^{-1}, indicating the plasma model with the same parameters used here can explain those results.

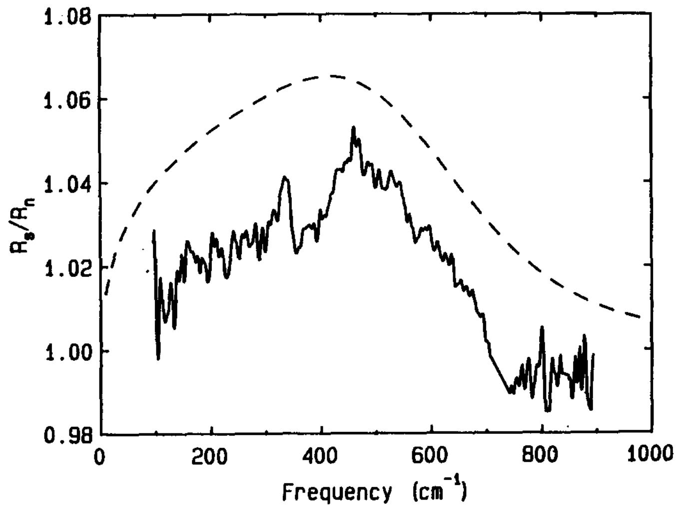

Fig. 4. The ratio of the reflectance in the superconducting state to the reflectance in the normal state at 100 K. This ratio would peak at the energy gap 2Δ in a BCS superconductor. Shown as a dashed line is a calculation that assumes the electrons have condensed to highly conducting state and that the gap is larger than $1/\tau$.

In summary, the normal-state properties are consistent with the combination of Drude and mid-infrared contributions to the conductivity. In the superconducting state, a plasma edge is seen in the far-infrared reflectance. No gap value can be inferred from the existing far infrared data.

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