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Mid-infrared reflectivity and ellipsometry measurements on single-crystal $YBa_2Cu_3O_7$ and $Bi_2Sr_2CuO_{6+y}$

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We have measured the room-temperature reflectivity of "90-K" single crystals $YBa_2Cu_3O_7$ in the frequency range 600–9000 cm^{-1}. The reflectivity R in these highly conducting samples ($\rho_{ab}=150$ $\mu\Omega$ cm at 290 K) is found to be higher than in previous reports. We fit R to a simple Drude-Lorentz model and compare the fit parameters with the dc transport values. (The effective-mass ratio of the carriers is found to be 2.0, and the scattering rate $\hbar/\tau$ is $3.1k_BT$ at room temperature.) Ellipsometry measurements have also been performed using transverse-electric and transverse-magnetic polarizations. The dielectric dispersion derived from ellipsometry shows some important deviations from the Drude-Lorentz model. Reflectivity data from nonsuperconducting crystals of $Bi_2Sr_2CuO_{6+y}$ are also reported.

I. INTRODUCTION

In the past two years many reports[1–8] have appeared on the infrared (ir) reflectivity of various high-T_c superconductors. The "90-K" superconductor $YBa_2Cu_3O_7$ (1:2:3) has been the most intensively investigated. Because of the large electronic anisotropy and the large variation of sample quality, however, there exists a wide range of measured reflectance. For example, the normal-state reflectivity R measured at the frequency 2000 cm^{-1} was reported to be 55% for ceramic samples of the "90-K" phase,[1] and 67% for an epitaxial thin film.[2] One of the first studies on single-crystal 1:2:3 reflectance was reported by the Tokyo group.[3,4] The many subsequent measurements of the reflectance of the a-b face of crystals have also shown a wide variation. At 2000 cm^{-1}, R was found to be 65% for a mosaic of single crystals of the "90-K" phase.[5] Crystals with a transition temperature $T_c=50$ K showed a reflectivity of 68%.[6] The highest reflectivity reported on a single crystal is 82%.[7] This disparity among different groups has greatly complicated the interpretation of the reflectivity data in 1:2:3. Since much important information on the electronic dynamics can be deduced from the frequency-dependent conductivity in the far-ir and mid-ir range, more accurate reflectivity data are obviously desirable.

A major goal of the present study is to determine the absolute reflectivity in single crystals of 1:2:3 at room temperature. By careful attention to sample quality, and instrumentation stabilization, we have achieved good reproducibility in determining the absolute reflectivity in a study extending to seven crystals of 1:2:3 in the "90-K" phase. R is reproducible to $\pm 1\%$ in a given crystal, and the disagreements between the three samples measured at near-normal incidence is less than 3%. The reflectivity obtained here is consistently higher than in previous reports, (by a factor of 2 at our highest frequency ω). We have compared the R-ω profile with the conventional Drude model and the Drude-Lorentz model (Sec. III). In spite of the high-reflectivity values reported here, we confirm that the simple Drude model remains wholly inadequate, as found in previous studies on samples with poorer reflectivity. By adding a Lorentz term to the Drude term (the "Drude-Lorentz" model) we significantly improve the fit to the data in our frequency range. Nonetheless, we have strong reservations about the physical validity of the parameters so obtained, primarily because of the ellipsometry results in Sec. IV. Some measurements on nonsuperconducting single crystals of $Bi_2Sr_2CuO_{6+y}$ (2:2:0:1) are also reported here, for comparison.

The ellipsometry measurements were performed on

1:2:3 with the angle of incidence fixed near 48°. By comparing R measured in the near-normal incidence mode with that in the transverse-electric (TE) and transverse-magnetic (TM) modes, we have derived the dispersion of the in-plane dielectric function ϵ_{ab} without resorting to a specific model. The dispersion of ϵ_{ab} shows the existence of a broad flat band of absorption between 4000 and 8000 cm^{-1} and a distinct break in slope in the conductivity centered at 3200 cm^{-1}. We show that the Drude-Lorentz model fails to account for this break which we believe to be intrinsic to the fully oxygenated phase of 1:2:3. Section V discusses our main conclusions and presents a comparison of our results with previous work.

II. EXPERIMENTAL DETAILS

The 1:2:3 crystals used were grown from a CuO-BaO flux at Princeton University. Several magnetization and transport measurements have been performed on these crystals.[9–11] All (as-grown) crystals investigated (by resistance or ac susceptibility) have shown a T_c consistently above 90 K, with a transition width ΔT_c from 0.1 to 0.2 K.[10] The field-cooled magnetization curves indicate a Meissner fraction of 13–15 % measured in a 100-G field perpendicular to the c axis.[11] The in-plane resistivity varies from 150 to 200 $\mu\Omega$ cm at room temperature.[10] The typical area of the largest face (a-b plane) is 1 mm^2. Micro-Auger spectroscopy studies on the a-b face show that, whereas the top 100–200 Å layer is deficient in Y and O (and rich in Cu and Ba), the bulk stoichiometry (particularly the relative oxygen content) remains unchanged to a depth of 5 μm. The uniformity and high quality of the crystals greatly decrease the overall uncertainties in the reflectivity measurements. Since long post annealing in oxygen (which degrades the surface quality) and culling a batch for the best T_c crystals are unnecessary, a tedious step in these experiments is eliminated. Much less characterization has been carried out for the new Bi 2:2:0:1 crystals. Resistance measurements show that they are not superconducting for T down to 4.2 K. Their reflectivity data are included here for comparison.

The infrared reflectance was measured in a commercial spectrometer specially aligned to minimize signal drift. The incident light, modulated by a rapid-scan interferometer and polarized by a Brewster angle polarizer, is focused on the a-b face of the sample, which is mounted at the end of a metal rod of diameter 0.8 mm. The beam spot size at focus is slightly larger than the sample area. A specially designed holder for the metal rod enables the sample to be removed and remounted easily without affecting the reflectance by more than 1%. The reflectivity of each sample is determined by taking the ratio of its reflectance with and without a gold film. The absolute reflectivity of the gold film was calculated for a given configuration from the optical data. We estimate that the overall uncertainty in the absolute reflectivity so determined is less than 1%. To minimize drift in the optics alignment, the instrument was allowed to stabilize for 1 d before actual measurements were made. The interferometer was operated in dry nitrogen-purge mode. All data are taken at room temperature. For the near-normal-incidence experiments the angle of incidence θ is between 11.6° and 12.6°, while for the ellipsometry measurements, θ is increased from 48.5° to 49.7°. In the latter experiments either the incident E field or B field is kept parallel to the a-b face (TE and TM modes, respectively).

Three samples of 1:2:3 (samples 1, 3, and 4) were studied at near-normal incidence. Two of them (3 and 4) came from the same growth batch. Sample 4 was measured two weeks after growth while sample 3 was measured four months after growth. The maximum difference in their reflectivity was 1.3%. As a further test of the correlation between R and the sample quality, we studied two samples (8,9) that were intentionally grown under off-stoichiometric conditions. These crystals, which are larger in area, have a higher in-plane resistivity (~400 $\mu\Omega$ cm at 290 K) and a lower T_c. They show a reflectivity significantly lower than samples 1–7 (~7% lower for $\omega < 7000$ cm^{-1}; 3–5 % for $\omega > 7000$ cm^{-1}).

III. FITS TO DRUDE AND DRUDE-LORENTZ MODELS

Figure 1 shows the reflectivity R of samples 1 and 2 in both TE and TM modes. In general, we find excellent reproducibility among the different samples up to 7000 cm^{-1}. The reflectivity of samples 5–7 lies between the curves of samples 1 and 2 for each polarization. The TM reflectivity of one sample (3) measured at $\theta = 49.7°$ showed 3–7 % higher reflectivity than the other five samples from 7000 to 9000 cm^{-1}. By rotating each sample by 90° about an axis normal to its a-b face, we are able to reproduce the R-ω profile to within 3% uncertainty. This indicates that the a-b plane is heavily twinned in all

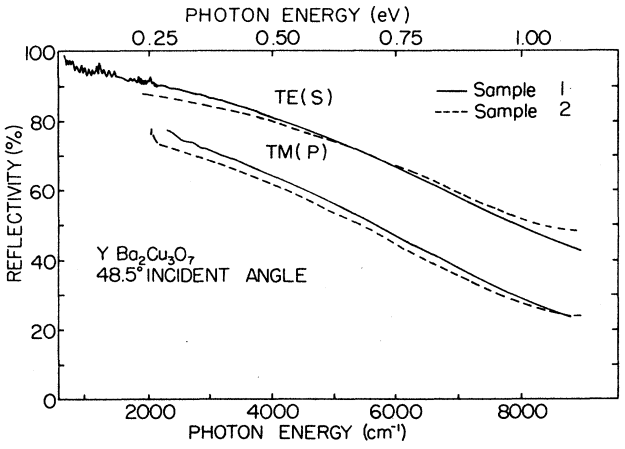

FIG. 1. Reflectivity vs frequency in the transverse electric (TE) and transverse magnetic (TM) modes for single-crystal $YBa_2Cu_3O_7$ (samples 1 and 2) at 290 K. The angle of incidence is 48.5°. The data for three other crystals (3–5) lie between the curves for these samples. The reflectivity of gold is assumed to be 100% for this figure.

our crystals, as confirmed by visual examination under polarized light.

Previous studies[1,5,6,8] have all shown that the simple Drude model fails to account for the reflectivity data in the far-ir and mid-ir range. We arrive at the same conclusion despite the fact that here R is substantially higher than in previous reports. Nonetheless, it is instructive to attempt a Drude-type fit to clarify where the major discrepancies lie, and the range of parameters that can be extracted. Figure 2 shows a fit to the near-normal-incidence reflectivity data using the Drude expression for the dielectric function (ω_p is the plasma frequency and τ the relaxation time)

$$\epsilon(\omega)=-\omega_p^2/[\omega(\omega+i/\tau)]+\epsilon_\infty \ . \tag{1}$$

The "best" fit [shown in Fig. 2(a)] is obtained with $\omega_p=20\,000$ cm^{-1} and $\tau^{-1}=3200$ cm^{-1}. For the large-θ data, similar parameters are obtained in the TE mode ($\omega_p=19\,000$ cm^{-1} and $\tau^{-1}=3100$ cm^{-1}) and in the TM mode ($\omega_p=19\,000$ cm^{-1} and $\tau^{-1}=2500$ cm^{-1}). (In all fits the high-frequency dielectric constant value, ϵ_∞, is fixed at 4.0 in accordance with previous work.[6,7] If we allow ϵ_∞ to vary, the fit is improved, but the fit to the TE data gives $\omega_p=36\,000$ cm^{-1}, $\tau^{-1}=4400$ cm^{-1}, and $\epsilon_\infty=21$. Even larger ω_p and ϵ_∞ are obtained for the near-normal incidence and TM data. We judge these numbers to be quite unrealistic in comparison with the available dc transport data. Also, the large ϵ_∞ would imply a highly polarizable ionic core.)

We note that the value of $\omega_p=20\,000$ cm^{-1} corresponds to a hole carrier density p equal to $4.5\times10^{21}(m^*/m_0)$ cm^{-1}, where m^*/m_0 is the hole effective mass normalized to the electron mass in vacuum. (From the Hall effect[12] p is estimated to be 1 hole/cell or 5.7×10^{21} cm^{-3}.) Using ω_p and τ ($=1.66\times10^{-15}$ s) extracted from the Drude fit, we compute a room-temperature in-plane dc resistivity $\rho_{ab}=1/(\epsilon_0\omega_p^2\tau)$ equal to 500 $\mu\Omega$ cm. This is to be compared with 150–200 $\mu\Omega$ cm measured directly. Thus, there are serious discrepancies between the Drude and dc transport values. The estimated p (for $m^*/m_0=1$) is comparable with the dc Hall data but the scattering rate is very high ($\hbar/\tau\sim15.4\ k_BT$), so that the in-plane conductivity is three times as small as the dc value. In view of the poor fit in Fig. 2(a) we will not discuss the Drude model further. [As shown in Fig. 2(a) the major manifestation of the poor fit is the pronounced downwards curvature of R calculated from Eq. (1). In contrast, all our results show a quasilinear decrease of R with increasing ω.]

In the single-plane Bi 2:2:0:1 crystals a similar linear variation or R versus ω is observed (Fig. 3). The quasilinear decrease of R in this range of ω may be a generic property of these oxides. It is also apparent in the published data (e.g., of the "50-K" phase of 1:2:3). Again, we have used Eq. (1) to attempt a fit (solid line) to R. The fit parameters yield a ω_p (15 600 cm^{-1}) much smaller than that for 1:2:3, but a τ^{-1} which is very similar (3300 cm^{-1}). With the caveats discussed above, we compare the Drude parameters with the dc transport numbers. The ratio p/m^* is only 60% as large as in 1:2:3. The Drude parameters imply a dc ρ_{ab} equal to 1200 $\mu\Omega$ cm (compared with 3000 $\mu\Omega$ cm measured by the Montgomery method[13]). Hall data are not available for this system.

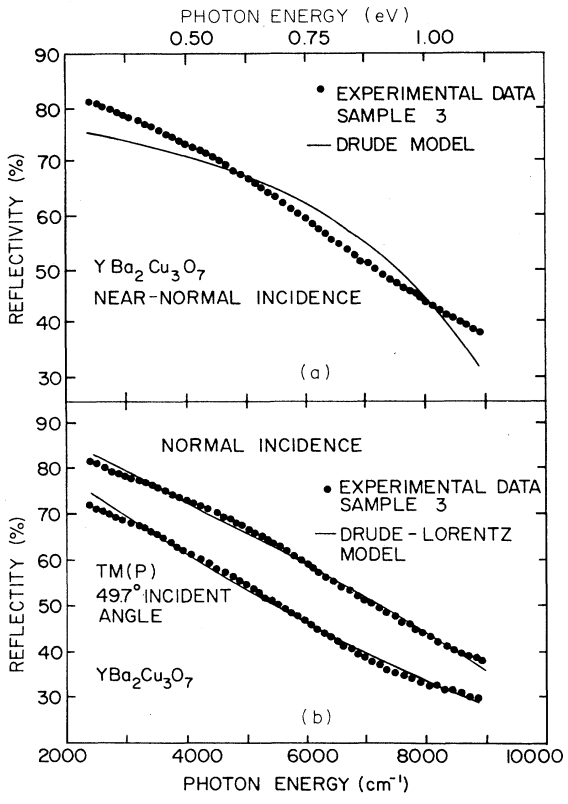

FIG. 2. (Top panel) fit of the Drude model [Eq. (1)] to the near-normal-incidence reflectivity for sample 3. (b) (Bottom panel) fits to the near-normal-incidence and TM mode reflectivities using the Drude-Lorentz model [Eq. (3)].

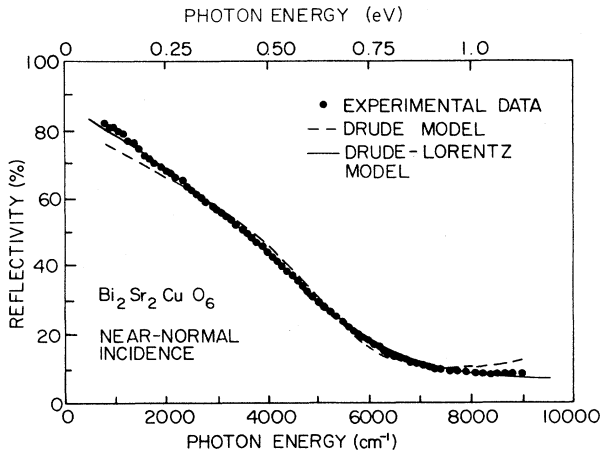

FIG. 3. The reflectivity of single-crystal $Bi_2Sr_2CuO_{6+y}$ at near-normal incidence (solid circles) and fits using the Drude model (dashed line) and the Drude-Lorentz model (solid line).

The pronounced deviations from the simple Drude behavior is most often ascribed to the existence of strong mid-ir absorption, for e.g., in the context of an excitonic mechanism for superconductivity.[1,8] In Fig. 2(b), we show a Lorentzian fit to the data given by

$$\epsilon(\omega)=-\omega_p^2/[\omega(\omega+i/\tau)] -\omega_{p1}^2/[(\omega^2-\omega_\gamma^2)+i\omega/\tau_1]+\epsilon_\infty \ . \quad (2)$$

In fitting the data to Eq. (2) we have imposed the constraints $\rho_{ab}=150\ \mu\Omega\,\mathrm{cm}$ and $\epsilon_\infty=4.0$. The parameters obtained from the near-normal-incidence data are $\omega_{p1}=21\,000\ \mathrm{cm}^{-1}$, $\tau_1^{-1}=11\,000\ \mathrm{cm}^{-1}$, $\omega_\gamma=2600\ \mathrm{cm}^{-1}$, $\omega_p=16\,000\ \mathrm{cm}^{-1}$, and $\tau^{-1}=640\ \mathrm{cm}^{-1}$. Fits performed separately on the TE and TM mode data yield similar values. From the new ω_p value, p is slightly smaller $[2.9\times10^{21}\ (m^*/m_0)]$. The new scattering rate is also much smaller ($\hbar/\tau=3.1\ k_BT$). In the literature, much smaller values of ω_p[3100,[6] 5900,[8] and 9700 cm^{-1} (Ref. 8)] have been obtained using Eq. (3). It is worth pointing out that, with the larger R measured here, it is not possible to obtain reasonable fits to Eq. (3) with ω_p smaller than 12 000 cm^{-1}. [With $\omega_p<12\,000\ \mathrm{cm}^{-1}$, the optimization procedure tends to force the resonance frequency ω_γ to zero, so that we revert back to Eq. (1)]. Thus, Eq. (3) appears to provide an adequate description in these high-reflectivity samples only if the strength of the Drude term is sufficiently large ($\omega_p>1.5$ eV). Even in this case, though, we have reservations about the Drude-Lorentz model because of the ellipsometry data (see the next section).

For the 2:2:0:1 crystals, we fitted the reflectivity with $\omega_{p1}=12\,000\ \mathrm{cm}^{-1}$, $\tau_1^{-1}=7100\ \mathrm{cm}^{-1}$, $\omega_\gamma=2000\ \mathrm{cm}^{-1}$, $\omega_p=8600\ \mathrm{cm}^{-1}$, and $\tau^{-1}=740\ \mathrm{cm}^{-1}$ with ϵ_∞ fixed at 4.0 (Fig. 3). (From these numbers we calculate a dc resistivity of 600 $\mu\Omega\,\mathrm{cm}$, whereas the measured resistivity equals 3000 $\mu\Omega\,\mathrm{cm}$.) A recent electron-energy-loss (EELS) experiment[14] on "two-plane" $Bi_2Sr_2CaCu_2O_8$ has found that ω_p is 1 eV in this oxide.

IV. DIELECTRIC FUNCTION FROM ELLIPSOMETRY

Ellipsometric measurements have been performed on 1:2:3 crystals in the visible to ultraviolet (uv) range.[15] As far as we know, such measurements have not been extended to the interesting mid-ir range. In principle, ellipsometry can be a powerful aid to the deconvolving the dielectric dispersion without assuming a particular model for the electronic dispersion. We have carried out a primitive version of ellipsometry using the near-normal incidence together with the TE and TM mode results at each ω.

First, we will briefly sketch our argument that because $|\epsilon_{ab}|$ is large we may ignore the out-of-plane dielectric function ϵ_c in analyzing the TM data. From the Fresnel equations for reflectivity of the TM mode the complex index of refraction $n(\theta_t)$ is given by

$$n(\theta_t)^{-2}=\epsilon_{ab}^{-1}\cos^2\theta_t+\epsilon_c^{-1}\sin^2\theta_t \ , \quad (3)$$

where the (complex) angle of transmission θ_t is related to the angle of incidence θ by Snell's law, $n(\theta_t)\sin\theta_t=\sin\theta$. For the values of ϵ_{ab} and ϵ_c measured we find that $|\sin^2\theta_t|\sim0.05$, so that we have, to leading order,

$$n(\theta_t)^{-2}=\epsilon_{ab}^{-1}[1+(\epsilon_c^{-1}\sin^2\theta-\sin^2\theta_t)] \ . \quad (4)$$

Because the magnitude of the term within ( ) is about 0.1, for our range of θ, more than 99% of R is determined by ϵ_{ab}. (This is the only approximation in the analysis.) Physically, the large values of $|\epsilon_{ab}|$ and $|\epsilon_c|$ (both >5) imply that the transmitted wave propagates almost parallel to the c axis, so that the in-plane dielectric response dominates the system's polarizability. Consistent with this, the Drude-Lorentzian fits to R yield very similar parameters when applied to the TM, TE, and near-normal-incidence measurements.

At each ω, we derived the real and imaginary parts, $\mathrm{Re}\epsilon_{ab}(\omega)$ and $\mathrm{Im}\epsilon_{ab}(\omega)$, by minimizing the sum of the squares of differences between calculated (subscript "calc") and measured (subscript "meas") reflectivities for each of the three modes,

$$(R_{\mathrm{TM\,calc}}-R_{\mathrm{TM\,meas}})^2+w(R_{\mathrm{TE\,calc}}-R_{\mathrm{TE\,meas}})^2 +(R_{N\,\mathrm{calc}}-R_{N\,\mathrm{meas}})^2 \ .$$

(The weight w is set between 0.3 and 0.6.)

An agreement of better than 1% was obtained between the calculated and measured reflectivities for ω up to 7000 cm^{-1}. The derived ϵ_{ab} below 7000 cm^{-1} is quite sensitive to the value of R, since a 1% disparity between say $R_{N\,\mathrm{calc}}$ and $R_{N\,\mathrm{meas}}$, was sufficient to change $\mathrm{Im}\,\epsilon_{ab}(\omega)$ by 20–60%. We have less confidence in the ellipsometric data beyond 7000 cm^{-1} because the fitting procedure is less satisfactory, and the sample-to-sample variation in R in the TM mode is considerable. Nonetheless, we note that both $\mathrm{Re}\epsilon_{ab}$ and $\mathrm{Im}\epsilon_{ab}$ derived here extrapo-

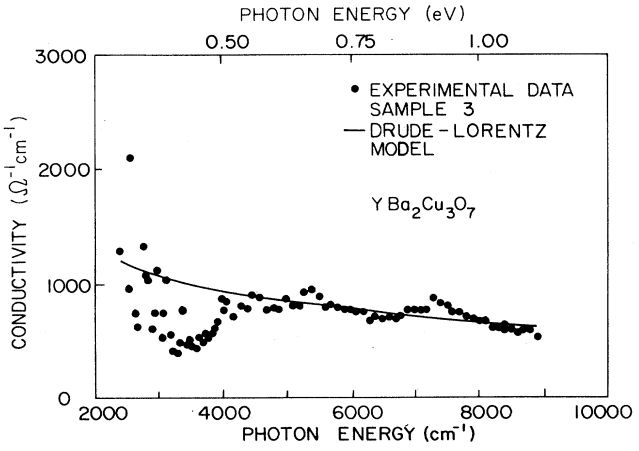

FIG. 4. Conductivity $\sigma(\omega)$ of $YBa_2Cu_3O_7$ estimated from ellipsometry using the near-normal-incidence reflectivity, and the reflectivity in the TE and TM modes. The solid circles are the conductivity determined ellipsometrically. The solid line is the Drude-Lorentz conductivity derived from fitting Eq. (2) to R.

late smoothly to the values obtained by conventional ellipsometry performed in the visible and uv range.[15]

The frequency dependence of the real part of the conductivity $\sigma(\omega)$ is displayed in Fig. 4. The most prominent feature, found in all our samples, is a broad and flat absorption band of strength $\sigma(\omega)\sim 1000\ \Omega\ \mathrm{cm}^{-1}$ between 4000 and 7000 cm^{-1}. Below 4000 cm^{-1}, $\sigma(\omega)$ shows a distinct break in slope near 3200 cm^{-1}. The Drude tail can be discerned below 3000 cm^{-1}, although the scatter in our fit is large in this range. For comparison, we show, as a solid line, the conductivity calculated from the fit in Sec. III to the Drude-Lorentz model [Eq. (2)]. Clearly, the fit fails to account for the break at 3200 cm^{-1}, although it describes $\sigma(\omega)$ fairly well above 4000 cm^{-1}. For the sample in Fig. 4 the break appears as a distinct minimum in $\sigma(\omega)$. However, data from other samples analyzed in a similar way show a break in slope at the same frequency range, rather than a minimum. At our level of resolution we can only state that a sharp change in $\sigma(\omega)$ occurs near 3000 cm^{-1}, and that this change cannot be described by Eq. (3).

V. DISCUSSION

As the quality and size of crystals improve, we can expect both an increase in the values of the reflectivity reported and convergence of the experimentally determined quantities. This trend is evident in various reports on single-crystal 1:2:3 in the past 18 months. In Fig. 5 we compare some of the published reflectivity data with the present work. At high $\omega(>8000\ \mathrm{cm}^{-1})$ the discrepancy between our results and some previous reports can differ by as much as 100%. At the low end ($<2000\ \mathrm{cm}^{-1}$) the difference in R is less pronounced. However, since the electronic parameters such as m^* and τ depend very sensitively on the difference $1-R$ (rather than on R) the discrepancy may be more serious. We have also displayed the data of Bozovic *et al.*[7] which were measured on 1:2:3 crystals grown at Princeton University. In general, the agreement with the present work is quite good. A recent study (Collins *et al.*[16]) also obtains R within a few percent of our values. This indicates that sample quality is the most important factor in determining R. Recent data by Thomas *et al.*[17] on a series of oxygen-depleted 1:2:3 crystals have also shown that R changes rapidly throughout the far-ir and mid-ir range as the oxygen content is lowered from 6.9 to 6.3.

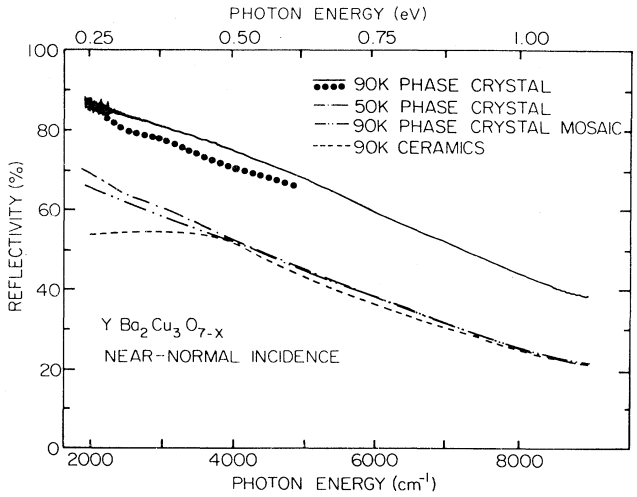

FIG. 5. A comparison of published reflectivity results on $YBa_2Cu_3O_7$ with the present work. From top to bottom at 2000 cm^{-1}, solid line (present work), solid circles (Ref. 5), dash-dot-dashed line (Ref. 4), dash-double dot-dashed line (Ref. 3), dashed line (Ref. 1). The reflectivity of gold is assumed to be 100% for this figure.

At best, we believe that the Drude-Lorentz model serves as a simple approximation to the conductivity spectrum. Approximating the flat absorption band between 4000 and 8000 cm^{-1} by a single Lorentzian is too crude, since clear deviations in the curvature of the measured R from the R calculated from Eq. (2) remains. Moreover, the break in slope in σ at 3200 cm^{-1} cannot be incorporated into Eq. (2). The actual spectrum in the mid-ir likely consists of several pieces. The two main components are a Drude-type term (for $\omega<1000\ \mathrm{cm}^{-1}$) and a broad absorption band above 4000 cm^{-1} which may be the sum of several Lorentzians. (This broad band may be related to the controversial 0.5-eV band first observed in ceramic samples.) Recent reports[6,16,17] have further clarified the structure at far-ir frequencies, and we refer the reader to them.

Since the effective mass m^* and scattering time τ of the carriers are of great interest in these systems, we discuss next the approximate values in 1:2:3 obtained from Eq. (2). Using the values $\omega_p=16\,000\ \mathrm{cm}^{-1}$ and $\hbar/\tau=640\ \mathrm{cm}^{-1}$, and assuming 1 hole/cell, we calculate an effective mass ratio $(m^*/m_0)=(\rho_{ab}pe^2\tau/m_0)$ equal to 2.0. This ratio is significantly smaller than previous estimates. If $\hbar/\tau$ at 290 K is actually smaller than our value (Thomas *et al.*[6] estimate $\hbar/\tau$ to be a factor of 2 smaller in their samples) m^*/m_0 will be slightly higher. However, an estimate larger than 4 would violate our fit to Eq. (2), or be inconsistent with the value of ρ_{ab}.

At high T, the relaxation time τ may be expressed in terms of an effective in-plane electron-phonon coupling constant λ_{ab} by the Bardeen relationship

$$\hbar/\tau=2\pi k_B T\lambda_{ab}\ . \tag{5}$$

The scattering rate $\hbar/\tau=3.1\ k_BT$ implies that $\lambda_{ab}=0.49$, which is a moderately large value compared with estimates from ceramic samples. A recent study[18] of the anisotropy of the thermal conductivity in single crystals also concludes that, although the out-of-plane electron-phonon coupling λ_c is negligible, the in-plane λ_{ab} is quite large.

VI. SUMMARY

We have measured the reflectivity from the a-b face of single-crystal $YBa_2Cu_3O_7$ in the mid-ir. For "90-K" crystals which have low in-plane resistivity (150 $\mu\Omega$ cm at 290 K), we have obtained a reflectivity spectrum which is accurate to 1–2 % for each sample. Below 7000 cm^{-1}, the sample-to-sample variation is less than 4%. We find

R to be significantly higher than in previous reports. A fit of R can be obtained using the Drude term plus a Lorentzian term [Eq. (2)]. Constraining the dc resistivity to the value measured on similar crystals, we find that the carrier relaxation rate $\hbar/\tau$ is 3.1 $k_B T$ and the effective mass ratio is 2.0. However, we have reservations about the validity of using a single Lorentzian to describe the rather broad absorption band starting at 4000 cm^{-1}, so that these numbers may require revision when more sophisticated models become available. From the ellipsometry data, we have also determined the dispersion of ϵ_{ab} independently of any model, and compared it with the fit using Eq. (2). With regard to the carrier dynamics, reflectivity data at ω lower than our range would be more helpful[16,17] (particularly at low T). However, the presence of the broad absorption band in the mid-ir range greatly distorts fits to the far-ir data if not treated correctly. We have attempted to minimize the uncertainty in the reflectivity results in the mid-ir. It is hoped that the spectral range and the strength of this broad band presented here may be helpful in analyzing the low-frequency results.

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