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OPTICAL CONDUCTIVITY OF THE HIGH-T_c's: SEARCH FOR THE ENERGY GAP

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ABSTRACT—The infrared absorption of oxide superconductors will be discussed. In the normal state, $\sigma_1(\omega)$ consists of a strongly T-dependent part in the far infrared, above which is a weakly-T-dependent midinfrared absorption. In the superconducting state, there is absorption to very low frequencies, well below where the superconducting gap is expected to be. It is argued that this absorption is due to a second ("midinfrared") component which is only weakly affected by the onset of superconductivity. The free carrier component is in the clean limit and most of its oscillator strength goes into the superfluid condensate zero-frequency delta function response.

Key words: High-T_c, infrared

INTRODUCTION

An ordinary superconductor has a gap Δ in its excitation spectrum. This gap causes the frequency-dependent conductivity $\sigma_{1s}(\omega)$ to be zero up to $\omega = 2\Delta$; above this frequency $\sigma_{1s}(\omega)$ rises to join the normal-state conductivity at several times 2Δ. The area removed from $\sigma_{1s}(\omega)$ appears under the zero-frequency delta function of the superconductor. The superconductor has zero absorption (100 % reflectivity) at frequencies below 2Δ and reduced absorption up to several times 2Δ.

Far-infrared reflection and transmission measurements were important in establishing the existence of a gap in metallic superconductors and in determining its magnitude. (Glover 1956, Ginsberg 1960, Palmer 1968, Drew 1967) With the discovery of the high-T_c compounds, there have been many attempts to do the same. (Timusk 1989, Tanner 1992) At the present time there is a lot of controversy—to say the least—about infrared determinations of the gap. This is a very complicated issue, and so we discuss it in some detail in the following paragraphs.

Very similar *ab*-plane reflectance spectra have been presented by a number of workers. (Timusk 1988, Thomas 1988, Schützmann 1989, Kamarás 1990, Renk 1990, Schlesinger 1990a, Cooper 1989, Orenstein 1990, Schlesinger 1987, Reedyk 1988, Collins 1989) In the superconducting state, the reflectance cannot be distinguished from 100% for $\omega \lesssim 140$ cm^{-1} (17 meV). Above 140 cm^{-1} there is a clear increase in absorption, and the reflectance drops to a plateau of about

98% which extends to a shoulder at ≈ 400 cm^{-1} (50 meV). At higher frequencies, the reflectance drops to join the normal-state data.

Both the 140 cm^{-1} onset and the 400 cm^{-1} shoulder have been assigned to the superconducting gap. Thomas *et al.*(1988) suggested that the apparent onset of absorption at ≈ 140 cm^{-1} in *ab*-plane data for reduced T_c samples might be the gap; this would give $2\Delta/k_BT_c = 3.2$. Schützmann *et al.*(1989) also put the gap at 140 cm^{-1} in measurements of 91 K T_c samples, which would make $2\Delta/k_BT_c = 2.1$

Schlesinger *et al.*(1987, 1990, Collins 1989) have interpreted the 400 cm^{-1} shoulder as the gap. When the ratio of superconducting to normal state reflectance, $\mathcal{R}_s/\mathcal{R}_n$, is plotted, this shoulder appears as a maximum. A maximum in $\mathcal{R}_s/\mathcal{R}_n$ does occur at 2Δ in ordinary superconductors because $\mathcal{R}_s$ is 100% out to 2Δ and then decreases to join $\mathcal{R}_n$, which has been decreasing like $1 - A\sqrt{\omega}$. This similarity to ordinary superconductors was the motivation for the $8k_BT_c$ assignment. (Schlesinger 1987, Collins 1989, Schlesinger 1990a, Schützmann 1989)

Data on untwinned crystals of $YBa_2Cu_3O_{7-\delta}$ show for $T = 30$ K an apparent 99–100% reflectance out to 500 cm^{-1} ($8k_BT_c$) for $\vec{E} \parallel a$ at which point there is a shoulder and decreasing reflectance. (Schlesinger 1990b, Rotter 1991) For $\vec{E} \parallel b$, the reflectance is smaller, with the shoulder less pronounced. Schlesinger *et al.*(1990) argue that the gap for *ab*-plane carriers must correspond to this onset and attribute the absorption below this frequency to carriers on the chains.

However, a more sensitive experiment than reflectance is direct bolometric absorption. Such experiments have been carried out by Pham *et al.*(1990, 1991) In untwinned crystals, they find a finite absorption down to their low frequency limit of 80 cm^{-1} ($1.2k_BT_c$) at all frequencies for both $E \parallel a$ and $E \parallel b$. The absorption for $E \parallel a$ is ~0.3% at 150 cm^{-1} and below. The direct absorption data are in agreement with reflectance data, but the better signal/noise ratio allows absorption to be seen at frequencies where the reflectance appears to reach 100%.

In this paper, we discuss the gap issue, using recent data on single-domain $YBa_2Cu_3O_{7-\delta}$ crystals, free-standing $Bi_2Sr_2CaCu_2O_8$ crystals, and $La_{2-x}Sr_xCuO_4$ thin films.

EXPERIMENTAL DETAILS

The single-domain crystals were prepared at the University of Illinois. The growth technique has been described previously. (Friedmann 1990) The crystals studied were about 2×2 mm in size and had extremely high quality surfaces. T_c was above 90 K. The reflectance was measured using Fourier and grating spectrometers and the optical constants determined using Kramers-Kronig analysis. (Quijada 1992) Estimated error in the far-infrared reflectance measurements, from reproducibility of different samples, is ±1–2%.

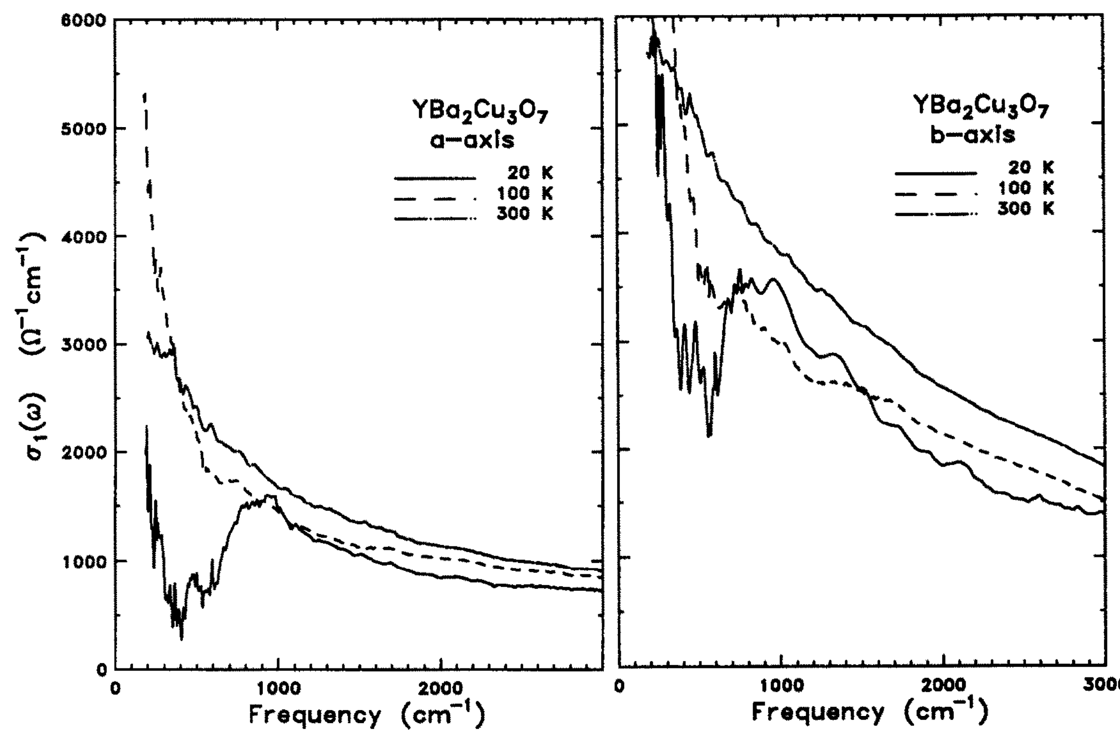
$La_{2-x}Sr_xCuO_4$ films were grown by off-axis magnetron sputtering on $SrTiO_3$ substrates at Westinghouse. Growth conditions and film properties have been described previously. (Talvacchio 1990) The film studied in detail was nearly 1 cm^2 in area, 8000 Å thick, and had good quality surfaces. X-ray analysis showed the film to be highly oriented with the *c*-axis normal to the surface. T_c was 31 K and x is ≈ 0.17. The reflectance was measured using Fourier and grating spectrometers and the optical constants determined using Kramers-Kronig analysis. (Gao 1992) No features attributable to the substrate could be seen in the spectra. Estimated error in the far-infrared reflectance measurements, from reproducibility of different samples, is ±0.5%.

Free-standing films (or flakes) of $Bi_2Sr_2CaCu_2O_8$ were prepared and characterized at SUNY Stony Brook as described in an earlier report. (Forro 1990) The samples were approximately 0.3 mm^2 in area and 1300–2000 Å thick. T_c was ≈ 82 K The transmittance was measured using Fourier and grating spectrometers and the optical constants determined using Kramers-Kronig analysis. (Romero 1991, Romero 1992a, Romero 1992b) Estimated relative error in the far-infrared transmittance measurements, from reproducibility of different samples, is ±5%. We note that transmittance is potentially more accurate than reflectance because an accurate 100% level is not required and that a 5% error in transmittance gives smaller errors in $\sigma_1(\omega)$ than a 0.5% error in reflectance. This is because the "signal" in the reflectance measurement is essentially $1 - \mathcal{R}$, and when $\mathcal{R} \to 1$ the signal goes to zero.

EXPERIMENTAL RESULTS

Fig. 1 shows the optical conductivity of a $YBa_2Cu_3O_{7-\delta}$ single crystal at three temperatures. The magnitude of the conductivity for $\vec{E} \parallel b$ is larger than for $\vec{E} \parallel a$ but otherwise the two spectra are similar and resemble the conductivity of twinned $YBa_2Cu_3O_{7-\delta}$ crystals (Orenstein 1990, Schlesinger 1990a) and films. (Schützmann 1989, Kamarás 1990) There is a strongly T dependent peak at low frequencies above T_c; below T_c a considerable amount of low-frequency oscillator strength is removed to the zero-frequency delta function conductivity of the superfluid. (That the oscillator strength moves to zero frequency and not, say, to some high energy can be proved by looking at the real part of the dielectric function. See Ref. Kamarás 1990 for an example.)

Below T_c there is a finite value of $\sigma_1(\omega)$ maintained to the lowest frequency measured (180 cm^{-1}) for both polarizations, in agreement with the results of Pham *et al.*(1991) (However, we should note that the uncertainties in the determination of $\sigma_1(\omega)$ are relatively large and reach nearly 100% at the low frequency limit of 180 cm^{-1}) A notch-like minimum is observed for both polarizations at around 500 cm^{-1}—above and below T_c—but no "gap" occurs in the spectrum at this frequency.



(a) (b)

Fig. 1. Optical conductivity of $YBa_2Cu_3O_{7-\delta}$ at three temperatures. (a) $\vec{E} \parallel a$. (b) $\vec{E} \parallel b$.

Fig. 2 shows the optical conductivity of $La_{2-x}Sr_xCuO_4$ at several temperatures. The inset shows (on a logarithmic frequency scale) the data into the visible region. As in the case of

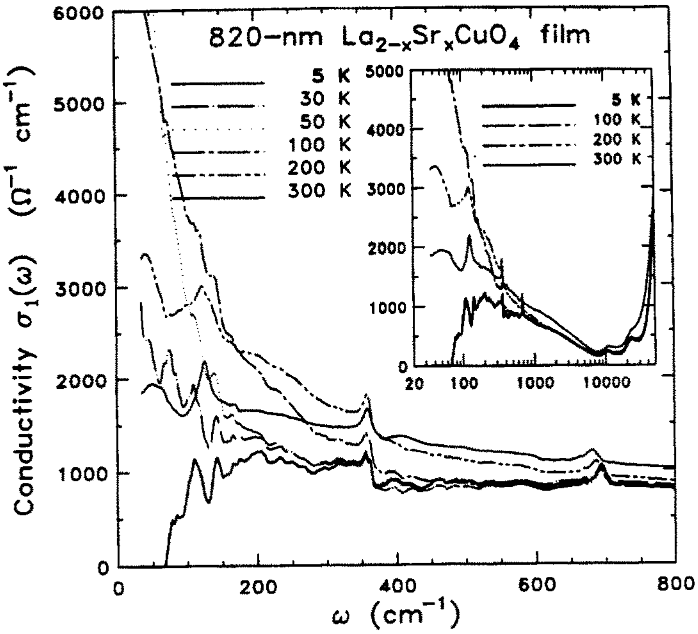


Fig. 2. Optical conductivity of $La_{2-x}Sr_xCuO_4$ at several temperatures. The inset shows the wide-frequency-range conductivity.

$YBa_2Cu_3O_{7-\delta}$, there is a strong T dependent peak at low frequencies. Below T_c a substantial amount of oscillator strength goes into the condensate.

The 5 K conductivity curve shows a threshold at 70 cm^{-1} but this is a consequence of the extrapolation; the uncertainty at 70 cm^{-1} is ±600 $\Omega^{-1}cm^{-1}$. This uncertainty is illustrated in Fig. 3, which shows the effect of scaling the reflectance up or down by the estimated uncertainty of $\pm0.5\%$. Propagated to $\sigma_1(\omega)$, this error becomes $\pm100\%$ by 100cm^{-1}. Another way of stating the same result is that our data is consistent with zero absorption below 100 cm^{-1} but not above 100 cm^{-1}.

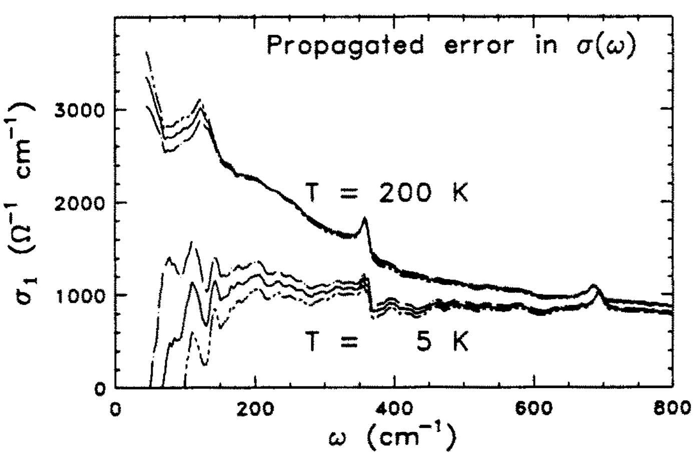


Fig. 3. Propagated uncertainty in $\sigma_1(\omega)$ from an uncertainty in reflectance of $\pm0.5\%$.

Fig. 4 shows the optical conductivity of $Bi_2Sr_2CaCu_2O_8$ at several temperatures. As in the other materials, the normal-state conductivity shows a strongly T-dependent upturn at low frequencies but minimal temperature dependence at higher frequencies. Below T_c, lots of

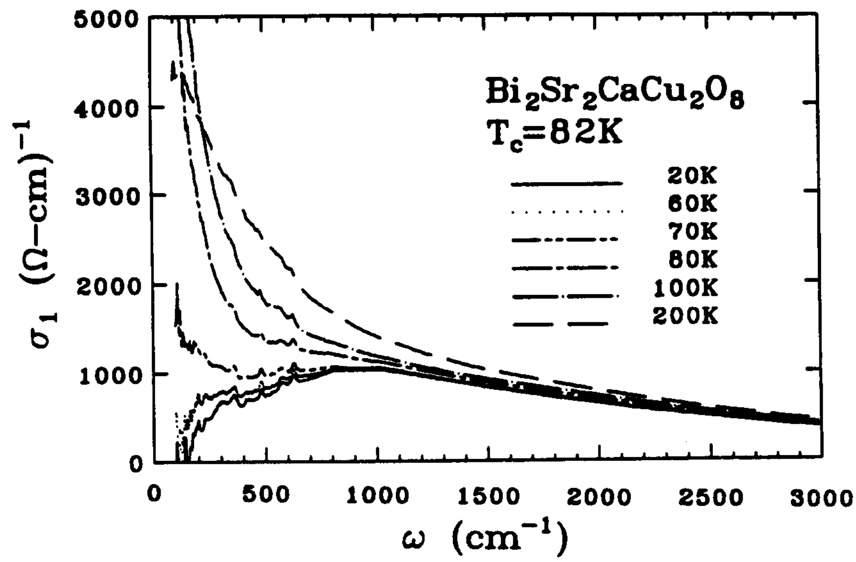


Fig. 4. Optical conductivity of $Bi_2Sr_2CaCu_2O_8$ at various temperatures above and below T_c.

oscillator strength moves into the condensate, but the conductivity is finite to 110 cm^{-1}. The error here is relatively small; there is definitely a positive value of $\sigma_1(\omega)$ at 140cm^{-1} and above.

DISCUSSION

We return now to the question of the superconducting gap. The results for thresholds in the optical conductivity are summarized in Table I. In all cases, the absorption threshold is well below $8k_BT_c$; only in $La_{2-x}Sr_xCuO_4$ would the data be consistent with a BCS gap value.

Table I. Energy scales in high T_c superconductors.

Material	Reference	T_c K	$3.5k_BT_c$ cm^{-1}	$8k_BT_c$ cm^{-1}	Absorption threshold cm^{-1}
$YBa_2Cu_3O_{7-\delta}$ $\vec{E} \parallel a$	This work	90	220	500	< 200
$YBa_2Cu_3O_{7-\delta}$ $\vec{E} \parallel a$	Pham *et al.* (1990)	90	220	500	< 80
$YBa_2Cu_3O_{7-\delta}$ twinned	Many. See Tanner and Timusk (1992).	92	230	511	< 140
$Bi_2Sr_2CaCu_2O_8$	This work	82	200	450	< 140
$La_{2-x}Sr_xCuO_4$	This work	31	75	170	< 100

This finite conductivity to low frequencies is difficult to interpret by analogy to conventional superconductors, where the picture of a superconducting gap works well. The difficulty is especially strong for $Bi_2Sr_2CaCu_2O_8$ where photoemission (Chang1989, Olson 1990) gives a rather convincing picture of a gap near $7k_BT_c$. As Fig. 4 shows, there is no particular anomaly in the spectrum at this frequency (400 cm^{-1}); indeed most of the fine structure in this region is also evident above T_c.

One resolution of this dilemma is to postulate that there are two contributions (or components) to the low-frequency optical conductivity: free carriers, which are responsible for the dc conductivity and which condense to form the superfluid below T_c, and bound carriers, which are relatively inert and which are not much affected by the superconducting transition. A wide variety of data have been analyzed in this picture. (Timusk 1989, Tanner 1992, Kamarás 1988, Bonn 1988, Timusk 1988, Thomas 1988, Schützmann 1989, Kamarás 1990, Renk 1890, Cooper 1989)

The dielectric function is then a sum free carriers (*i.e.*, a Drude model) and the midinfrared bound carrier terms,

$$\epsilon(\omega) = \epsilon_{MIR} - \frac{\omega_{pD}^2}{\omega^2 + i\omega/\tau} + \epsilon_\infty \tag{1}$$

where ω_{pD} is the plasma frequency of the free carriers and $1/\tau$ is their (essentially ω-independent) relaxation rate. The T-linear temperature dependence of the resistivity is assumed to come from the temperature dependence of $1/\tau$ since in the Drude model, $\rho = 4\pi/\omega_{pD}^2\tau$. The bound carriers are in a broad, nearly T-independent, band throughout the midinfrared.

The midinfrared contribution to the data of Fig. 4 is illustrated by Fig. 5. A Drude function has been subtracted from the total conductivity in order to obtain these curves. The Drude contribution has a plasma frequency of 10,300 cm^{-1}, a T-linear scattering rate, and agrees well with the dc conductivity. (Romero 1992) The mid-infrared contribution is almost independent of temperature above T_c, in agreement with hypothesis, but there are some systematic variations around 0.1 eV. Below T_c the Drude contribution has condensed, revealing more clearly the midinfrared band.

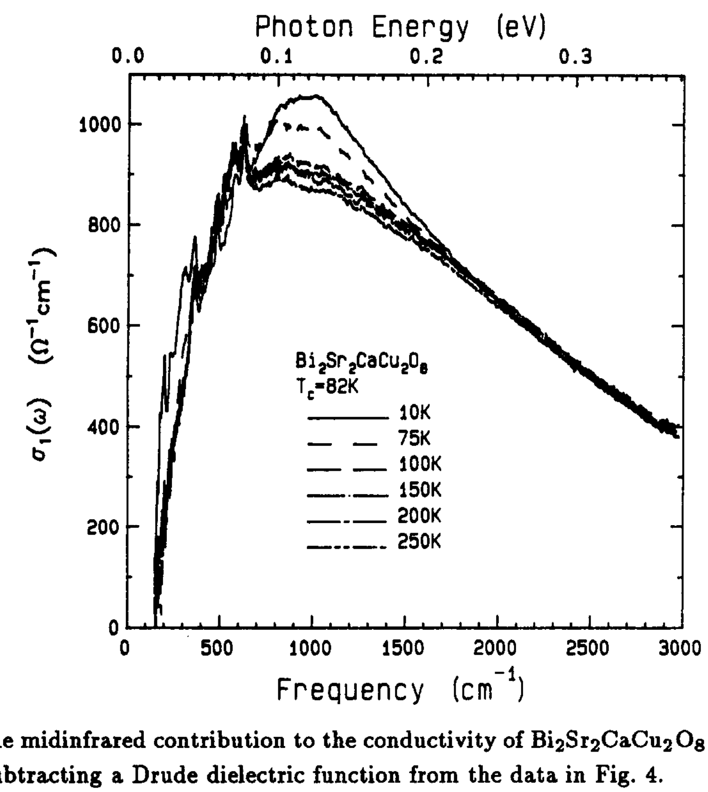


Fig. 5. The midinfrared contribution to the conductivity of $Bi_2Sr_2CaCu_2O_8$ crystals, obtained by subtracting a Drude dielectric function from the data in Fig. 4.

If the two-component picture of the midinfrared absorption is correct, then the presence of the second component can obscure any gap absorption and make its determination difficult. This effect occurred some time ago in early studies of $La_{2-x}Sr_xCuO_4$ ceramics, which showed a sharp reflectance drop in the 50 cm^{-1} region. This initially was assigned to the gap, but it later was shown to be caused by a zero-crossing of $\epsilon_1(\omega)$, due to an interplay between the negative contribution of the superfluid and a strong positive contribution from a phonon. (Bonn 1987, Sherwin 1988) Using a similar approach, Timusk *et al.*(Timusk 1988) argued that the maximum seen in $\mathcal{R}_s/\mathcal{R}_n$ in $YBa_2Cu_3O_{7-\delta}$ was affected by dispersion as well.

Kamarás *et al.*(1990) showed that various features in the superconducting-state conductivity spectrum of $YBa_2Cu_3O_{7-\delta}$ which have been assigned to the gap can also be seen in the normal-state conductivity. (This is also evident in the $Bi_2Sr_2CaCu_2O_8$ data of Fig. 5, where the total conductivity below T_c resembles closely the midinfrared contribution above T_c.) Because the features persist above T_c, it was argued that they were from the non-Drude midinfrared absorption and not associated with the superconducting gap. Above T_c this absorption is partially masked by the absorption of the free carriers; below T_c, when the free carriers condense into a delta function, the midinfrared absorption becomes fully revealed.

The question naturally arises: why should the gap not seen? Kamarás *et al.*(1990) suggested that this is because the high-T_c materials are in the "clean limit," with $2\Delta \gg 1/\tau$. In this limit, all of the free-carrier oscillator strength exists at low frequencies and goes into the zero-frequency delta function conductivity of the superconductor. None is left for transitions across the gap. The presence of the midinfrared absorption is a key point in the clean limit argument. Given sufficient sensitivity, the gap can be seen even in the clean limit of ordinary metals with no other low energy excitations. However, in the presence of the midinfrared absorption, especially if there is some temperature dependence in it, picking out the gap becomes more difficult.

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