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To cite this article: D. B. Tanner , M. A. Quijada , D. N. Basov , T. Timusk , R. J. Kelley , M. Onellion , J. P. Rice , D. M. Ginsberg , B. Dabrowski , S-W. Cheong , F. C. Chou & D. C. Johnston (1996) The far-infrared conductivity of oxide superconductors, *Ferroelectrics*, 177:1, 83-94, DOI: [10.1080/00150199608216955](https://doi.org/10.1080/00150199608216955)

To link to this article: <http://dx.doi.org/10.1080/00150199608216955>



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THE FAR-INFRARED CONDUCTIVITY OF OXIDE SUPERCONDUCTORS

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(Received June 26, 1995)

A number of open questions remain about the infrared response of high- T_c superconductors. Although there is clear evidence for the formation of a superconducting condensate, there is no convincing data showing a superconducting gap absorption in the far infrared spectrum. Most of their spectral weight of the free carriers goes into the superconducting condensate in the superconducting state. Most cuprates are orthorhombic crystals, so there is anisotropy in their transport and optical properties. In $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, the anisotropy of the London penetration depth shows that the chains contribute strongly to the superfluid. In $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$, where chains are absent, there is still a definite anisotropy to the far-infrared absorption, with a finite absorption for $\mathbf{E} \parallel \mathbf{b}$ down to ~ 20 meV. This anisotropy of the ab plane could be due either to anisotropy of the superconducting gap or to anisotropy of the midinfrared component to the conductivity.

Keywords: Superconductors, optical properties, high- T_c .

1. INTRODUCTION

The low-energy electronic excitations in the cuprate superconductors are due to the charge carriers introduced onto the CuO_2 planes by doping. In the undoped state,

the materials are generally agreed to be charge-transfer insulators,¹ with a gap of 1.5–2 eV. Upon doping, there is a significant transfer of oscillator strength to low frequencies and a corresponding reduction of the strength of the charge-transfer excitation.^{2–5} The optical conductivity in the low energy region has a number of unusual behaviors.^{6–30} The normal-state temperature and frequency dependence differs from that of ordinary metals. There is evidence for interaction of an unusual type between the charge carriers and certain optical phonons.¹⁹ In some cases, there are clearly two components to the conductivity spectrum, a free-carrier part and a bound-carrier (or ‘‘midinfrared’’) part, which act in parallel. In other cases, the two components are not so obvious and one can analyze the data in terms of strong frequency dependence to the quasiparticle self-energy function. In the superconducting state, there is clear evidence for the existence of a condensate but no convincing evidence for a superconducting gap absorption has been presented.

In this paper, the low frequency optical conductivity of three cuprate superconductors will be presented and compared: $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$, and $\text{La}_2\text{CuO}_{4+\delta}$. All three of the materials studied are close to their optimal doping level. All three are orthorhombic, so that the optical conductivity is a tensor quantity, with three different principal values, corresponding to polarization along the three crystallographic directions. This anisotropy is a topic for this paper.

Optical measurements are sensitive only to certain types of anisotropy, both in normal and superconducting states. Basically, quantities like Δ , the gap, must have only a twofold axis of symmetry in the ab plane. Then, the a and b components of the dielectric tensor will differ. If the ab plane has a fourfold axis, then the optical properties will be isotropic. Thus, although the optical conductivity is affected by the anisotropic order parameter of an unconventional superconductor,³¹ in not every case does the anisotropy of the order parameter lead to an anisotropic ab -plane optical conductivity. As an example, the optical conductivity for pure $d_{x^2-y^2}$ pairing is isotropic in the ab plane (although the spectrum *is* different from the case of s -wave pairing). In contrast, a p -wave component, a d_{xz} pairing, or a combination of s - and d -symmetries can give anisotropic ab -plane optical conductivities.

2. EXPERIMENTAL DETAILS

The samples studied were single crystals. The methods for growing these crystals and their properties have been described in detail elsewhere. Reference 32 describes the preparation and characterization of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals. Briefly, the crystals were grown using a self-flux technique in yttria-stabilized zirconia crucibles. The formation of twins was avoided by rapid cooling through the tetragonal-orthorhombic transition temperature. This results in tetragonal crystals which could be removed from the growth flux and subjected to a post-growth oxygen anneal to convert them to high- T_c superconductors. The post-growth anneal was carried out in flowing oxygen with the crystals supported on a wafer of polycrystalline $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. These single-domain crystals have zero resistance at 92 K and a dc resistivity anisotropy greater than 2.1 between 100 and 275 K. The extrapolation of both a - and b -axis resistivities to zero temperature gives nearly zero intercept.

The single crystals of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ used in this study were grown using slow-

cooling in a temperature gradient, as described by Han *et al.*³³ Typical crystals are thin rectangular platelets with a surface area of a few mm² in the *ab*-plane. Four-probe resistance gives a linear resistivity (with nearly zero intercept) in the normal state, zero resistance at typically 85 K, with a transition width of ~2 K. The principal axes were identified from the extinction points when rotating the sample in a microscope with crossed polarizers; they correlate with the *a* and *b* directions as observed with LEED, where *b* is the superlattice direction. Note that in the Bi₂Sr₂CaCu₂O₈ structure the *a* and *b* axes are along the Bi—O bonds, nearly 45° from the Cu—O bonds.

The La₂CuO_{4+δ} sample was prepared by electrochemical insertion of oxygen into La₂CuO₄. This technique allows the synthesis of uniformly oxidized samples with relatively high oxygen content ($\delta \sim 0.12$) and transition temperatures near 40 K.^{34,35} The process began with a single crystal of La₂CuO₄, prepared using a self-flux method.³⁶ The oxidation of this crystal was carried out using an electrochemical cell with the La₂CuO₄ sample as the working electrode.^{35,37} The cell was charged by applying an anodic current of 10 μ A to the La₂CuO₄ crystal for a period of two months. Meissner effect data indicate an onset of superconductivity at around 40 K.³⁷

Experimental considerations limited us to measuring only two of the three principal directions for each of our samples. In the case of La₂CuO_{4+δ}, the *ab* plane was twinned; however, the crystal was sufficiently thick that we could measure both $E\|ab$ and $E\|c$. The Bi₂Sr₂CaCu₂O₈ and YBa₂Cu₃O_{7-δ} crystals were single domain, so that the *a* and *b* axes independently could be measured, but the samples were in the form of very thin plates, so that there was not enough area in the faces containing the *c* axis to allow reflectance measurements to be performed.

The polarized optical reflectance was measured at several temperatures over ~80–40,000 cm⁻¹ (10 meV–5 eV). A Perkin-Elmer 16U grating spectrometer was used in the near-infrared to ultraviolet regions. The far-infrared and midinfrared regions were covered using a Bruker IFS 113v Fourier transform spectrometer. The temperature of the sample was varied by using a flow cryostat with a calibrated Si-diode thermometer mounted nearby. Determination of the absolute value of the reflectance was done by coating the sample with a 2000 Å film of Al after measuring the uncoated sample. The spectra of the uncoated sample were then divided by the obtained spectrum of the coated sample and corrected for the known reflectance of Al. The accuracy in the absolute reflectance is estimated to be $\pm 1\%$.

3. OPTICAL CONDUCTIVITY

The real part of the conductivity, $\sigma_1(\omega)$, was obtained from a Kramers-Kronig analysis of the reflectance.³⁸ The usual requirement of the Kramers-Kronig integrals to extend the reflectance at the low- and high-frequency ends was done in the following way. At low frequencies, the extension was done by modeling the reflectance using a Drude-Lorentz model and using the fitted results to extend the reflectance below the lowest frequency measured in the experiment. The high-frequency extrapolations were done by merging the data with results from the literature³⁹ or by using a weak

power law dependence, $\mathcal{R} \sim \omega^{-s}$, with $s \sim 1-2$. The highest frequency range was extended with a power law $\mathcal{R} \sim \omega^{-4}$, which is the free-electron behavior limit.

Figures 1-3 show respectively the optical conductivity of $\text{La}_2\text{CuO}_{4+\delta}$, $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$, and $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. Data are shown in both normal and superconducting states and for both polarizations measured.

a. $\text{La}_2\text{CuO}_{4+\delta}$

The left panel of Figure 1 shows the *ab*-plane optical conductivity of the $\text{La}_2\text{CuO}_{4+\delta}$ crystal and the right panel shows the *c*-axis conductivity. The anisotropy between the two polarization directions is evident in the figure. The *ab*-plane spectrum has considerable low-energy spectral weight whereas the *c* axis spectrum shows only the response of optical phonons. There are phonons at 230, 340, 492, and 512 cm^{-1} . The appearance of two phonons around 500 cm^{-1} , where only one is expected,⁴⁰ suggests that the incorporation of extra oxygen atoms in the structure splits the apical oxygen stretch into two modes.⁴¹ The electronic contribution to the *c*-axis conductivity is extremely small, indicating the highly two-dimensional behavior of this compound. Note that this quasi-two dimensional behavior exists, even though the system has "metallic" dc properties and becomes superconducting around 40 K. Similar effects have been observed in other systems.^{42,43}

The *ab*-plane conductivity of $\text{La}_2\text{CuO}_{4+\delta}$ has the following properties. (1) The temperature dependence at frequencies above 500 cm^{-1} occurs between 200 and 300 K; below 200 K the temperature dependence is small. This temperature dependence

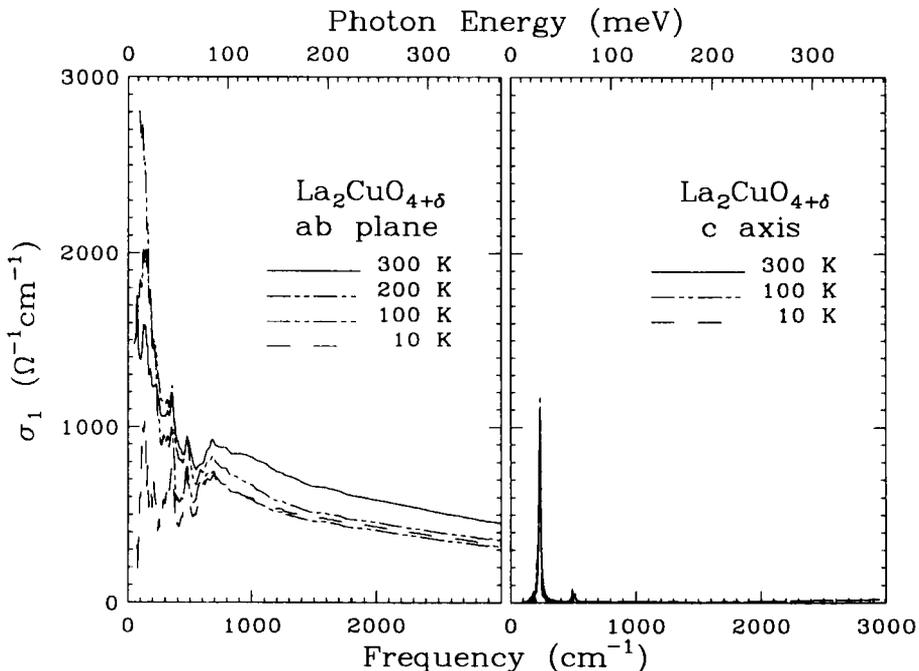


FIGURE 1 Optical conductivity of $\text{La}_2\text{CuO}_{4+\delta}$. Left panel: $\mathbf{E} \parallel ab$ plane. Right panel: $\mathbf{E} \parallel c$.

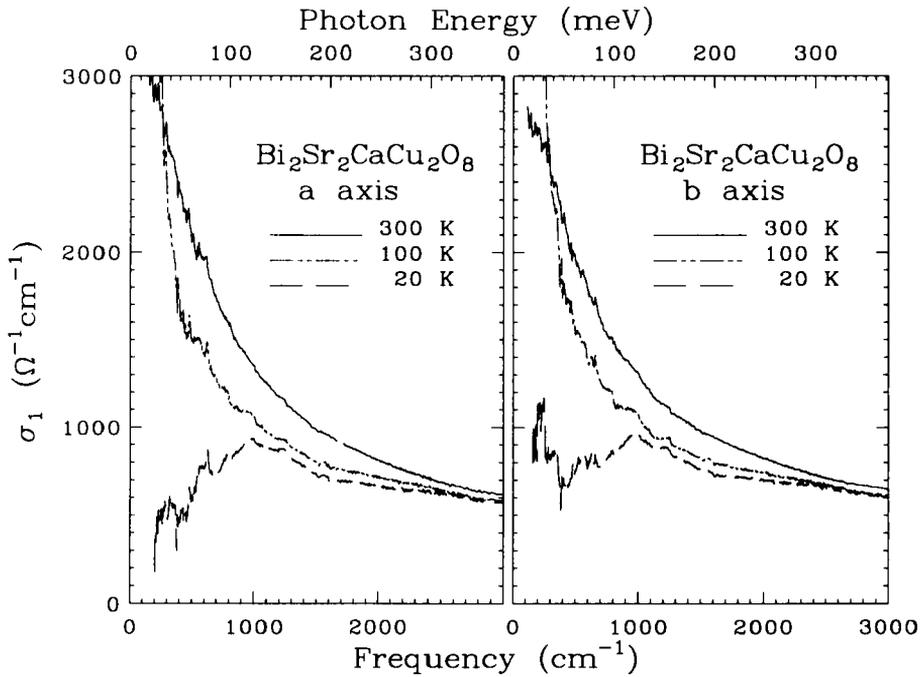


FIGURE 2 Optical conductivity of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$. Left panel: $\mathbf{E}||\mathbf{a}$. Right panel: $\mathbf{E}||\mathbf{b}$.

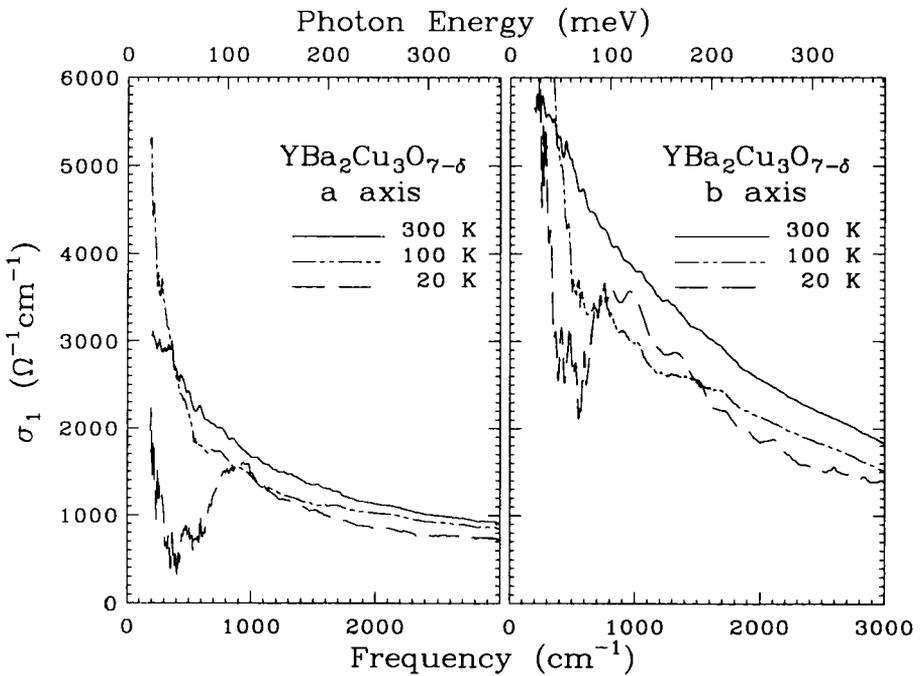


FIGURE 3 Optical conductivity of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. Left panel: $\mathbf{E}||\mathbf{a}$. Right panel: $\mathbf{E}||\mathbf{b}$.

differs from that of the dc resistance and is thought to be related to either ordering of the excess oxygen atoms or a growth in the local orthorhombic order in this temperature range.^{25,41} (2) In the normal state there is an increase in $\sigma_1(\omega)$ at the lowest frequencies which is in accord with the dc resistivity. The conductivity below 300–400 cm^{-1} is approximately Drude-like, a zero-frequency peak which grows and sharpens as temperature is reduced. (3) Weak phonon features are seen in the spectrum at 80, 140, 230, 355, 484, and 690 cm^{-1} . Three of these (140, 355, and 690 cm^{-1}) can be assigned to the E_u vibrations of La_2CuO_4 while the other three may be Raman modes activated by the disorder associated with the doping. (4) There are minima or “notch-like” structures in the frequency range 450–500 cm^{-1} , which result from electron-phonon interaction as has been discussed in detail elsewhere.^{19,44} (6) Below T_c , there is a considerable transfer of oscillator strength from the finite-frequency, far-infrared region to the zero-frequency, delta-function response of the superconductor. The oscillator strength of the delta-function is essentially the same as that of the Drude-like zero-frequency peak of the normal state. (7) There is a lot of low-energy oscillator strength remaining below T_c . A sum-rule evaluation finds that about 20% of the total doping-induced oscillator strength is in the delta function; the remainder is in the infrared spectrum shown for $T = 20$ K. (8) At all temperatures, there is a weak maximum at 700 cm^{-1} in $\sigma_1(\omega)$. This maximum suggests that the conductivity must be considered to consist of two components, a free-carrier part and a bound-carrier or “midinfrared” part. The free carrier part may have somewhat of a non-Drude character, resulting from a frequency dependent damping and effective mass.^{45–49} Alternatively, if a weak-coupling approach is justified, a simple Drude model is sufficient to describe the free-carrier part.^{6,7,15,41}

b. $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$

The left panel of Figure 2 shows the *a*-axis optical conductivity of a $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ crystal and the right panel shows the *b*-axis conductivity. The anisotropy between the two polarization directions is small, being most evident in the 20 K data. However, the *a*-axis conductivity is larger than the *b*-axis conductivity in the normal state. (The opposite is the case in the near-infrared-ultraviolet.³⁰) This anisotropy in the optical conductivity is consistent with the dc resistance of these samples.⁵⁰

The *a*- and *b*-axis conductivities of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ have the following properties. (1) The temperature dependence at frequencies above 1000 cm^{-1} is relatively modest; it is in fact mostly due to a narrowing of the Drude-like peak at zero frequency. The temperature dependence is much weaker than would be suggested by the dc resistance. (2) In the normal state there is an increase in $\sigma_1(\omega)$ at the lowest frequencies in accord with the dc resistivity. The conductivity below 300–400 cm^{-1} is approximately Drude-like, a zero-frequency peak which grows and sharpens as temperature is reduced. (3) Weak phonon features are seen in the spectrum, not notably at 390, 480, and 610 cm^{-1} for the *a*-axis polarization and 400, 630 and 650 cm^{-1} for the *b*-axis polarization. We believe that these are the ordinary in-plane vibrations of the system. (4) There is a minimum or “notch-like” structure in the frequency range 450–500 cm^{-1} . (6) Below T_c , there is a considerable transfer of oscillator strength from the far-infrared region to the delta-function response of the superconductor. The oscillator strength of the delta-function is essentially the same as that of the Drude-

like zero-frequency peak of the normal state. (7) There is a lot of low-energy oscillator strength remaining below T_c . A sum-rule evaluation finds that about 20–25% of the total doping-induced oscillator strength is in the delta function; the remainder is at finite frequencies. (8) Below T_c , there is a weak maximum at 1000 cm^{-1} in $\sigma_1(\omega)$. This maximum is not seen in the normal state; however, if a Drude curve is subtracted from the normal-state data, then the remainder, or “midinfrared” part, has this maximum.

That there is anisotropy in the optical conductivity of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ is consistent with its orthorhombic crystal structure. However, the anisotropy in the superconducting state is surprising. Generally one tends to view $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ as almost tetragonal, with a fourfold axis of symmetry about the copper site, and with the other structural elements less important for the superconducting state. Then, the differences between the a and the b directions in the superconducting state can arise in one of two ways. If there is only one component to the infrared conductivity, as in a marginal Fermi liquid,⁴⁵ nested Fermi liquid,⁴⁷ or other models,⁵¹ then the anisotropy reflects a two-fold symmetry to the superconducting gap absorption. (There is no other low-lying absorption band in these pictures.) As mentioned above, this anisotropy would be inconsistent with a purely $d_{x^2-y^2}$ gap symmetry. The second possibility is that there is a second component to the optical conductivity, so that the anisotropy could be attributed to this second component. (This “midinfrared” absorption must exist in order to assign the observed anisotropy to it.)

c. $\text{YBa}_2\text{Cu}_3\text{O}_{7-8}$

The left panel of Figure 3 shows the a -axis optical conductivity of a $\text{YBa}_2\text{Cu}_3\text{O}_{7-8}$ crystal and the right panel shows the b -axis conductivity. Note that the vertical scale is compressed by a factor of two compared with Figures 1 and 2. The anisotropy between the two polarization directions is about a factor of two at all temperatures.

The a - and b -axis conductivities of $\text{YBa}_2\text{Cu}_3\text{O}_{7-8}$ have the following properties. (1) There is a modest temperature dependence at high frequencies, mostly due to a narrowing of the Drude-like peak at zero frequency and much weaker than suggested by the dc resistance. (2) In the normal state there is an increase in $\sigma_1(\omega)$ at the lowest frequencies, in accord with the dc resistivity. It is approximately Drude-like, a zero-frequency peak which grows and sharpens as temperature is reduced. (3) Weak phonon features are seen in the spectrum, but they are not much larger than the noise level. We believe that these are the ordinary in-plane vibrations of $\text{YBa}_2\text{Cu}_3\text{O}_{7-8}$. (4) There is a strong minimum or “notch-like” structure in the frequency range $450\text{--}500\text{ cm}^{-1}$. This can be seen in the data below T_c very clearly and is also in the 100 K data.^{15,19} (6) Below T_c , there is a considerable transfer of oscillator strength from the far-infrared region to the delta-function response of the superconductor. The oscillator strength of the delta-function is essentially the same as that of the Drude-like zero-frequency peak of the normal state. (7) There is a lot of low-energy oscillator strength remaining below T_c . A sum-rule evaluation finds the delta function consumes about 20–25% of the total doping-induced oscillator strength for both polarizations, even though the area under the delta function for $\mathbf{E}\parallel a$ is only about half that for $\mathbf{E}\parallel b$. (8) Below T_c , there is a weak maximum at 1000 cm^{-1} in $\sigma_1(\omega)$. This maximum is not seen in the normal state; however, if a Drude curve is sub-

tracted from the normal-state data, then the remainder, or “midinfrared” part, has this maximum.

There is about a factor of two difference in the total doping-induced oscillator strength for the two polarizations. This anisotropy is consistent with the transport anisotropy in the normal state.³² The dc resistivity anisotropy in the *ab*-plane of YBa₂Cu₃O_{7- δ} , ρ_a/ρ_b , is 2–2.3. Using a two-component analysis of the normal-state optical conductivity,⁷ we find a similar anisotropy in the normal state Drude plasma frequency $\omega_D^2 = (4\pi ne^2)/m^*$: $\omega_{Db}^2/\omega_{Da}^2 = (16,000 \text{ cm}^{-1}/10, 500 \text{ cm}^{-1})^2 = 2.2 \pm 0.2$. These normal state results are in good agreement with LDA band structure calculations⁵² which have predicted an anisotropy close to 2.3 for the plasma frequency of YBa₂Cu₃O_{7- δ} . This anisotropy is a consequence of three bands that cross the Fermi surface, one of which is principally chain-like and has a highly anisotropic effective mass.

4. SUPERCONDUCTING PENETRATION DEPTH

Optical data can be used to obtain the principal components of the penetration depth tensor.⁵³ Methods commonly used to probe the superfluid, such as muon spin resonance (μ SR), yield values of λ_L which average the components of the penetration depth tensor. DC magnetization and microwave methods, while yielding the temperature dependence of the penetration depth with great accuracy, generally cannot be used to obtain absolute values; these methods also involve a mixture of at least two of the tensor components in a given measurement.

The superconducting penetration depth λ_L can be found from the reflectance in two ways. From the Kramers-Kronig analysis of one may obtain the imaginary part $\sigma_2(\omega)$ of the complex conductivity as well as the real part $\sigma_1(\omega)$. $\sigma_2(\omega)$ is directly related to λ_L via

$$c^2/\lambda_L^2(\omega) = \omega_{ps}^2 = 4\pi\omega\sigma_2(\omega), \quad (1)$$

where ω_{ps} is plasma frequency of the condensate given by $4\pi n_s e^2/m^*$. Here, n_s is the superfluid density and m^* the effective mass of the carriers. Equation 1 is a direct consequence of the delta function peak in the real part of the conductivity at $\omega = 0$. Alternatively, the strength of delta function peak can be obtained from the oscillator strength sum rule,

$$c^2/\lambda_L^2 = 8 \int_0^\infty (\sigma_{1n} - \sigma_{1s}) d\omega, \quad (2)$$

where σ_{1n} and σ_{1s} are the real parts of the optical conductivity in normal and superconducting states. Equation 2 states that spectral weight lost at low frequencies in the superconducting state has been transferred to the zero frequency delta function response of the superconducting condensate.⁵⁴

If $\lambda_L(\omega)$ in Equation 1 is ω -independent then both Equations 1 and 2 may be used to estimate λ_L . This is the case for our data. The values of the penetration depth obtained by these two methods agree within about 10%. Figure 4 shows the frequency dependent penetration depth (top to bottom, in order of increasing transition

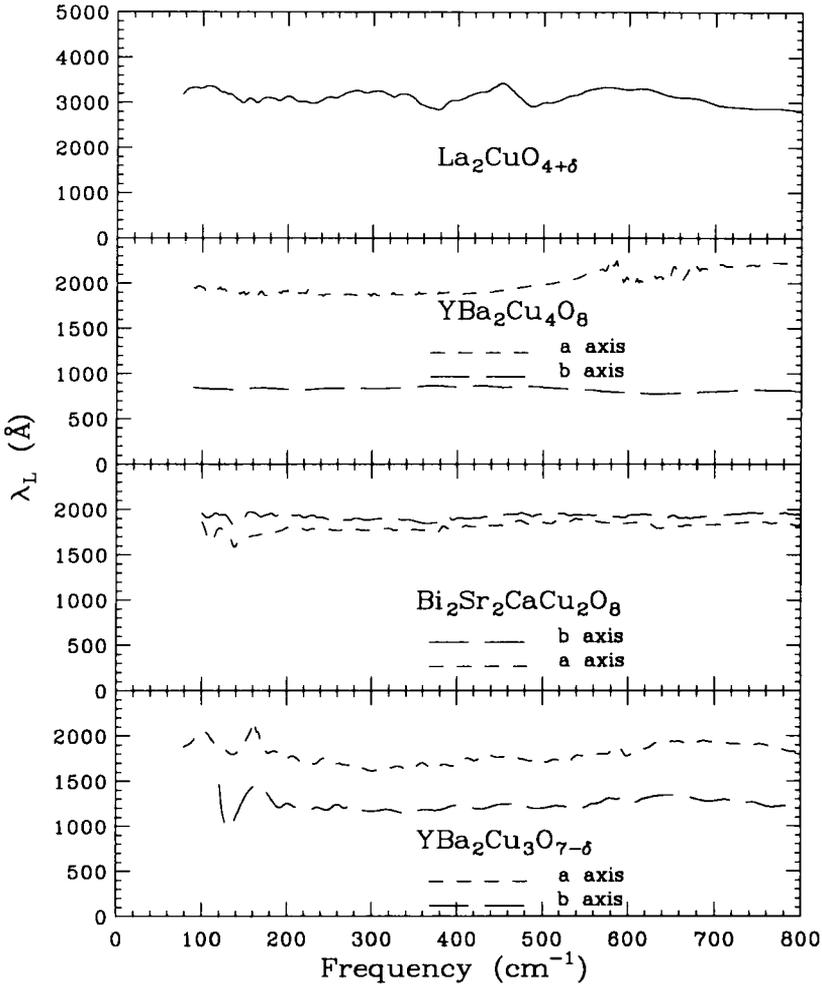


FIGURE 4 The frequency-dependent penetration depth of four cuprate superconductors. Upper panel: $\text{La}_2\text{CuO}_{4+\delta}$. Second panel: $\text{YBa}_2\text{Cu}_4\text{O}_8$. Third panel: $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$. Bottom panel: $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. The sample temperatures were 10 K, 10 K, 20 K, and 20 K, respectively.

temperature) for $\text{La}_2\text{CuO}_{4+\delta}$, $\text{YBa}_2\text{Cu}_4\text{O}_8$, $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$, and $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. Note that a smaller λ_L would correspond to a larger superfluid density. For the chain-containing materials ($\text{YBa}_2\text{Cu}_4\text{O}_8$ and $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$) the correlation between superfluid density and transition temperature^{55,56} holds only if the *a*-axis penetration depth is used.

The superconducting penetration depth in untwinned single crystals of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and $\text{YBa}_2\text{Cu}_4\text{O}_8$ is smaller in the chain direction than it is normal to the chains; therefore, a large portion of the spectral weight that can be attributed to the chains in the normal state condenses below T_c . These results indicate that superconductivity, at least in these compounds, is not confined to the planes but extends to the chains as well. For $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, the anisotropy of the penetration depth is remarkably close to the anisotropy of the normal state conductivity, the Drude plasma frequency, and the LDA calculations. Thus, the value of $(n/m^*)_b/(n/m^*)_a$ taken from

any of these experiments is 2.2 ± 0.2 . The chains not only increase the spectral weight in the normal state but also enhance the superfluid density by the same factor. This suggests that the condensate resides on both *the chains and the planes*. In qualitative agreement with this picture, the anisotropy of both the normal state properties and of λ_L is even more pronounced in the double chain 124 system.⁵³

5. CONCLUSIONS

The optical properties of the three materials discussed here have many similarities but also some differences. Among these is the increase in low-energy oscillator strength from $\text{La}_2\text{CuO}_{4+\delta}$ to $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ to $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and a corresponding increase in the plasma frequency and the superfluid density inferred from the penetration depth. A second difference is the greater anisotropy within the *ab*-plane for the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ material as compared to $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$. However, the anisotropy in the latter material is not negligible.

The anisotropy in λ_L in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (and $\text{YBa}_2\text{Cu}_4\text{O}_8$) implies that there is a large superfluid density on the chains. One may imagine then that all of the sheets of the Fermi surface participate in the superconductivity. Despite this result, the optical conductivity in the superconducting state shows no sign of a gap, such as exists in ordinary metallic superconductors. This absence of a gap can be explained in one of two ways: a clean-limit¹⁵ argument in a two-component picture or the presence of an unconventional gap³¹ with nodes somewhere on the Fermi surface. The anisotropic optical conductivity in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ is inconsistent with a pure $d_{x^2-y^2}$ symmetry for this gap, although other possibilities, discussed above, exist.

ACKNOWLEDGEMENTS

Work supported at the University of Florida by NSF Grant No. DMR-9403894, at McMaster by the Canadian Institute for Advanced research (CIAR) and by the National Science and Engineering Research Council (NSERC), at Wisconsin by NSF grant DMR-8911332, at the University of Illinois and at Northern Illinois University by NSF Grant DMR 91-20000 through the Science and Technology Center for Superconductivity. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under contract No. W-7405-ENG-82. The work at Ames was supported by the Director for Energy Research, Office of Basic Energy Sciences.

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