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$12k_BT_c$ optical signature of superconductivity in single-domain $Bi_2Sr_2CaCu_2O_8$

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Infrared and far-infrared transmittance measurements were performed on $Bi_2Sr_2CaCu_2O_8$ (2:2:1:2) and $Bi_2Sr_2CuO_6$ (2:2:0:1). Below T_c, a characteristic feature occurs at $\omega=700$ cm^{-1} ($12k_BT_c$) in the 2:2:1:2 material. No similar feature is found in the 2:2:0:1 compound. The 2:2:1:2 material also showed substantial *ab*-plane anisotropy.

The nature of the low-frequency optical excitations in the high-temperature superconductors is still controversial and unresolved.[1] Reflectance measurements of *ab*-plane oriented samples consistently have shown two reflectance edges at low frequencies, each being a possible candidate for the superconducting gap.[2–8] A value of $2\Delta=3.5k_BT_c$ was proposed by Thomas *et al.*,[2] and $2\Delta=8k_BT_c$ was suggested by Schlesinger *et al.*,[7,9] while Kamarás *et al.*[6] argued that the gap in the quasiparticle density of states may not lead to observable optical features at all. The superconducting gap, if it is indeed observed, is highly unusual since its magnitude would not depend on the temperature[4,6] and only slightly on T_c.[4] The normal state properties are also unusual: the frequency dependence of the optical conductivity is clearly different from simple Drude response.[1–9]

In this paper, we present infrared transmittance of the superconducting $Bi_2Sr_2CaCu_2O_8$ (2:2:1:2) compound ($T_c=82$ K) and the nonsuperconducting (but metallic) $Bi_2Sr_2CuO_6$ material ($T_c<3$ K) in the frequency range of 100–3000 cm^{-1} (0.01–0.37 eV) and for temperatures between 7 and 300 K. The measurements were made on free-standing single crystals.[10] We observe a characteristic, temperature-dependent optical feature around $\omega\approx700$ cm^{-1} (i.e., $\hbar\omega\approx12k_BT_c$) in the 2:2:1:2 material but not in the 2:2:0:1 compound. Measurements with polarized radiation revealed that the *ab*-plane optical properties of the 2:2:1:2 compound are anisotropic.

The preparation and characterization of the samples is described in an earlier report.[11] The crystals were approximately 0.3 mm^2 in area and 1300–2000 Å thick with the *c* axis along the short direction. Selected area electron diffraction shows an incommensurate superlattice along the *b* direction of the 2:2:1:2 samples. The samples also proved to be untwinned, i.e., the superlattice structure has the same direction throughout the sample area and thickness. The clearly distinct diffraction patterns allow us to exclude the presence of a second phase or 2:2:0:1 inclusion in the 2:2:1:2 sample.

We measured the absolute transmittance of our free-standing single crystals at temperatures from 7 to 300 K. For metallic samples, the transmittance $\mathcal{T}$ has the advantage of being less sensitive than the reflectance $\mathcal{R}$ to systematic errors. Crudely speaking, this is because $\mathcal{T}$ is measured relative to 0% while $\mathcal{R}$ is relative to 100%, and it is hard to determine with great accuracy the 100% reference. We used beamline U4-IR at the National Synchrotron Light Source[10] over 100–700 cm^{-1} and a Bruker fast-scan Fourier transform spectrometer, globar source, and 4 K Si:B photodetector over 400–3000 cm. $^{-1}$. The estimated error in our measurements is $\delta\mathcal{T}=\pm0.0005$ below about 2000 cm^{-1}, increasing to ±0.005 at higher frequencies. Since the signal transmitted by the sample is much weaker than the reference signal, we checked the linearity of the photodetector response with the intensity of the incident radiation. To minimize the effects of drifts in the spectrometer, sample and reference spectra were taken at each temperature.

The unpolarized transmittance of the oriented 2:2:1:2 crystal is shown in Fig. 1 (upper panel) along with the normalized transmission, $\mathcal{T}_T/\mathcal{T}_{250K}$ (lower panel). The inset shows the ratio for sample 2. Below T_c, the transmittance extrapolates to zero at $\omega=0$. This behavior, a general property of superconductors, is essentially a consequence of the screening of electromagnetic fields by the superfluid and has been observed on other *ab*-plane oriented samples of $Bi_2Sr_2CaCu_2O_8$ (Ref. 10) and $YBa_2Cu_3O_{7-\delta}$.[12] The low-frequency transmittance in the normal state increases with *T*, in accord with the dc resistivity. At higher frequencies (above 500 cm^{-1}), however, the temperature dependence of the transmittance is reversed, i.e., the transmittance decreases with increasing temperature. There are weak phonon resonances at 480 and 650 cm^{-1}.

In the superconducting state, the transmittance develops a characteristic "shoulder" around 700 cm^{-1}; this feature is clearly visible for each sample but is somewhat sharper for sample 2. In the normalized transmission

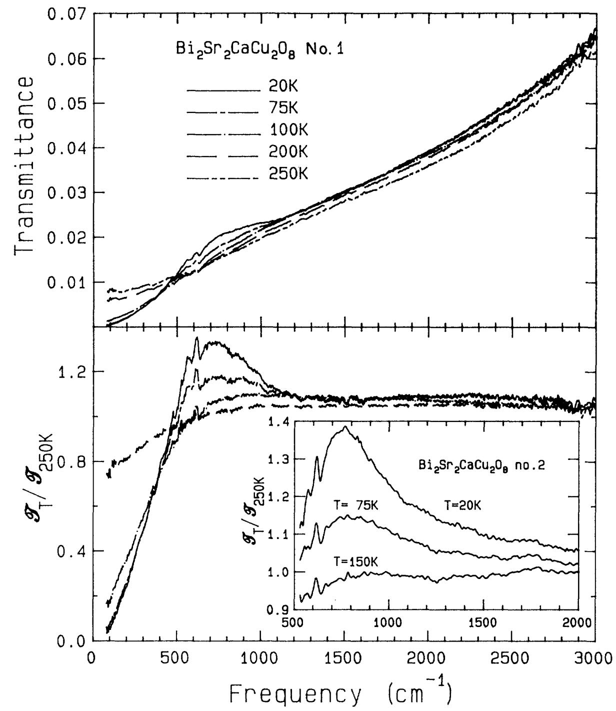

FIG. 1 Upper panel: transmittance spectra of a 1350-Å thick $Bi_2Sr_2CaCu_2O_8$ sample at temperatures between 20 K and 250 K. Lower panel: The ratio, $\mathcal{T}_T/\mathcal{T}_{250K}$, of the spectra shown above. The inset shows $\mathcal{T}_T/\mathcal{T}_{250K}$ for a second 2:2:1:2 sample.

(lower panel of Fig. 1) this feature is a peak, which appears only below T_c.

The results for the 2:2:0:1 sample are shown in Fig. 2. Although the transmittance is temperature dependent, no particular structure appears at any temperature. The temperature dependence of the low-frequency limit, again, follows from the temperature dependent dc resistivity. No crossover of the transmittance is observed for this sample. The normalized transmission (Fig. 2 inset) exhibits a gradual downturn at low frequencies but no peak is seen. Thus, the comparison between the 2:2:1:2 and 2:2:0:1 samples clearly indicates that the peak around 700 cm^{-1} is a unique property of the superconducting samples.

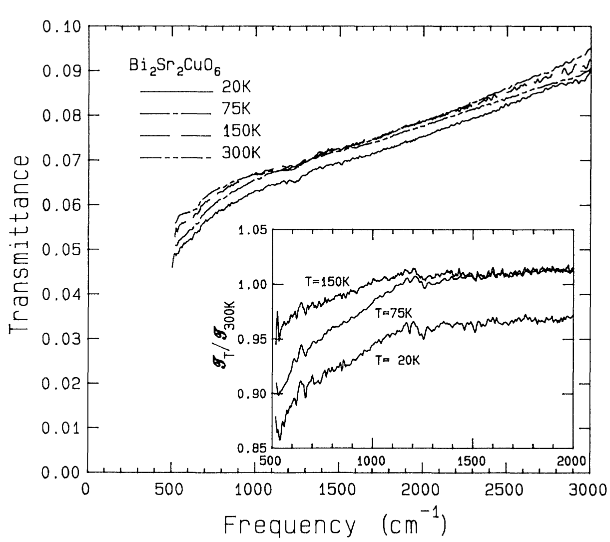

FIG. 2 Transmittance of a nonsuperconducting $Bi_2Sr_2CuO_6$ sample at temperatures between 20 K and 300 K. The inset shows $\mathcal{T}_T/\mathcal{T}_{300K}$.

The polarization dependence of the 20-K transmittance for 2:2:1:2 crystal 1 is shown in Fig 3. In order to exclude instrumental problems, we recorded spectra both with fixed sample, rotating the polarization of the light, and with fixed polarization, rotating the sample, we obtained identical results in both cases. For $\mathbf{E}\|a$, the transmittance ($\mathcal{T}_a$) was larger than for $\mathbf{E}\|b$ ($\mathcal{T}_b$) at every temperature and frequency. At 1000 cm^{-1}, the difference $(\mathcal{T}_1-\mathcal{T}_b)/\mathcal{T}_a$ is about 12%; this decreases to 10% at 150 K. The 700-cm^{-1} shoulder is present for both polarization, indicating that it has no relation to the b-axis superlattice. The phonon resonance at 650 cm^{-1}, on the other hand, is stronger for $\mathbf{E}\|b$ where it acquires an asymmetric Fano line shape.

An in-plane anisotropy of the visible-region spectrum was observed by Kelly *et al.*[13] while an anisotropy of the dc resistivity was reported by Martin *et al.*[14] Thus, it is clear that within the ab plane the electronic properties of this pseudotetragonal compound are anisotropic. Recent measurements on untwinned $YBa_2Cu_3O_{7-\delta}$ samples[9] reveal ab anisotropy as well. In Ref. 9, this difference is assigned to the chains and the anisotropy in the 2D CuO_2 layers is taken to be zero. Our results on 2:2:1:2—which has no chains—indicate that this cannot be the entire story.

Our present state of understanding of the optical conductivity of high-temperature superconductors offers two, fundamentally different, interpretations for the data. The first, based on frequency dependent relaxation rates in the normal state and the opening of a superconducting

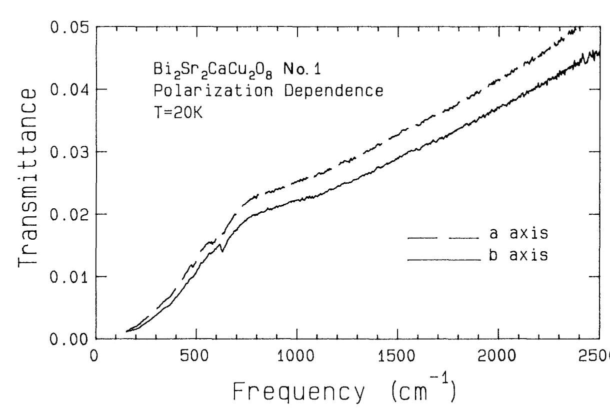

FIG. 3 Polarized transmittance of a single-domain $Bi_2Sr_2CaCu_2O_8$ sample at 20 K.

gap below T_c,[2,7,9] assumes only one type of charge carrier. Coupling of these carriers to some (optically inactive) excitations leads to a frequency dependent relaxation rate and effective mass. The "marginal Fermi liquid" (MFL) theory of Varma *et al.*[15] and the "nested Fermi liquid" (NFL) theory of Virosztek and Ruvalds[16] provide a phenomenological background for this approach. The second interpretation assumes two components: a low-frequency Drude conductivity plus optically active excitations in the midinfrared.[17,18,2,4,6,10] In this case, the superconducting gap may not be observable at all on account of the materials being "clean-limit" superconductors.[6]

We found that both models result in adequate fits to our data. We determined the temperature dependence of the parameters in each model.

In the MFL or NFL model, the self-energy (Σ) of the free charge carriers is frequency dependent:[15,16,19] at low frequencies, $-\mathrm{Im}\ \Sigma \sim \pi\lambda_T T$, while at high frequencies, $-\mathrm{Im}\ \Sigma \sim \lambda\omega$ up to a cutoff frequency, ω_c. The dielectric function is $\epsilon(\omega)=\epsilon_\infty - \omega_p^2/\omega(\omega-\Sigma)$. To facilitate our fits we used an expression for Σ with the appropriate analytical properties:

$$\Sigma(\omega)=\frac{2\lambda\omega}{\pi y}\ln\left[\frac{\pi T y - i\omega}{\omega_c y - i\omega}\right] - i\pi\lambda_T T \ , \qquad (1)$$

where $y=1-(\omega/\omega_0)^2$ and ω_c and ω_0 are characteristic cutoff frequencies. For $\pi T \ll \omega_c < \omega_0$ and for frequencies $\omega < \omega_c$, Im Σ satisfies the criteria put forward by Varma *et al.*[15] For $\omega < \pi T$, the optical response is that of a Drude metal with relaxation rate, $1/\tau = \pi\lambda_T T$; for $\pi T < \omega < \omega_c$, the effective relaxation rate is approximately linear in frequency.

Using the expression of Eq. (1) we found that we could fit our data well, with the following results. (1) The plasma frequency $\omega_p = 14\ 100 \pm 200\ \mathrm{cm}^{-1}$. (2) The coefficient of the T-linear term $\lambda_T = 1.53 \pm 0.02$. (3) The coefficient λ of the ω-linear term was temperature dependent, decreasing by about a factor of 2 between 100 and 300 K. Furthermore, it differed from the coefficient of the T term. (4) The cutoff frequencies $\omega_c = 1200 \pm 100\ \mathrm{cm}^{-1}$ and $\omega_0 = 1800 \pm 100\ \mathrm{cm}^{-1}$. Note that, especially at higher temperatures, the true "marginal Fermi liquid" limit (i.e., $\pi T \ll \omega_c$) is thus not realized. With these caveats, the MFL model works quite well in the normal state.

For the two-component model, the midinfrared contribution was represented by a single Lorentzian oscillator (with the parameters of oscillator strength, ω_{pe}, center frequency, ω_e, and width, γ_e). From dc resistivity, $\rho = 4\pi/\omega_{pD}^2\tau$, where ω_{pD} is the plasma frequency of the free carriers, we concluded that the normal-state relaxation rate follows $\tau^{-1} = 1.7T$. In the superconducting state, the free carrier response had a superfluid component, $A\delta(\omega)$, as well, with a relative weight, A, increasing at lower temperatures. At $T \simeq 75$ K, approximately 60% of the total Drude spectral weight went into the delta function, with complete condensation to the superfluid at $T \le 50$ K.

The two-component model also gave good fits to the data. The two components had nearly equal and nearly T-independent strengths, $\omega_{pe} = 11\ 000 \pm 200\ \mathrm{cm}^{-1}$ and $\omega_{pD} = 10\ 100 \pm 200\ \mathrm{cm}^{-1}$; the total integrated oscillator strength in the infrared, ($\omega_{pD}^2 = \omega_{pe}^2$) is within 5% of that from the MFL fit. Although the midinfrared contribution had a modest temperature dependence (most notably the center frequency of the midinfrared band increased from 800 ± 50 to $900 \pm 50\ \mathrm{cm}^{-1}$ and its width decreased as the temperature is lowered) we may conclude that the main features of the measured transmittance can be described with the superposition of a free carrier and a midinfrared contribution to the response, where the dominant temperature dependence is carried by the linear variation of the free-carrier relaxation rate.

We now turn to the peak seen in the transmittance below T_c. The peak seen in the $Bi_2Sr_2CaCu_2O_8$ films is consistent with other measurements on high-T_c superconductors. As an example, we show in Fig. 4 the transmittance for a hypothetical 1350-Å film of $YBa_2Cu_3O_{7-\delta}$, calculated from the reflectance data of Kamarás *et al.*[6] These $YBa_2Cu_3O_{7-\delta}$ data predict a maximum in $\mathcal{T}_s/\mathcal{T}_n$ (inset) at 550 cm^{-1}, a little below the maximum in $Bi_2Sr_2CaCu_2O_8$. The maximum correlates nicely with a local minimum in the reflectance below T_c.

The observation of a peak in $\mathcal{T}_s/\mathcal{T}_n$ near $\omega = 2\Delta$ in conventional superconductors provided strong support for the energy gap model and the validity of the BCS theory of superconductivity.[20] The situation in the high-T_c superconductors is not so simple.

The frequency dependent conductivity for a superconductor with the MFL model has recently been worked out by Nicol *et al.*[21] The most important result is that for the clean limit (which in this context means that the impurity scattering is small compared to 2Δ), the optical absorption begins at 4Δ not at 2Δ.[22] The reason is that for absorption to occur, the photon energy must be sufficient (a) to break a Cooper pair (2Δ) and (b) to create a charge or spin fluctuation (also 2Δ), for a total of 4Δ. If the 700-cm^{-1}($12k_BT_c$) peak is associated with the gap

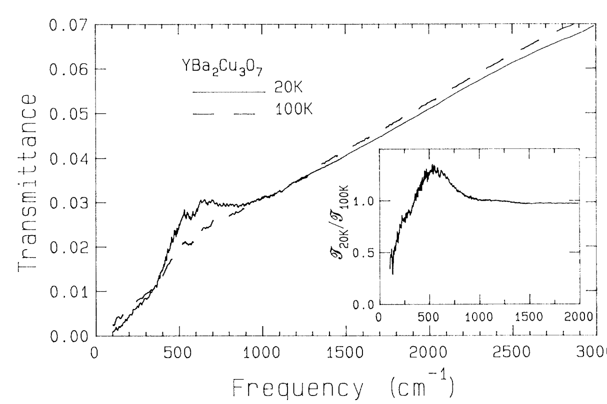

FIG. 4 Transmittance of a 1350-Å thick $YBa_2Cu_3O_{7-\delta}$ film at 20 and 100 K, calculated from the Kramers-Kronig derived optical constants of Ref. 6. The inset shows $\mathcal{T}_{20\ K}/\mathcal{T}_{100\ K}$.

in an MFL, then $2\Delta = 350\ \mathrm{cm}^{-1} = 6k_B T_c$. In addition, the calculations of Nicol *et al.*[21] imply that the $8k_B T_c$ feature of Schlesinger *et al.*[7,9] in $YBa_2Cu_3O_{7-\delta}$— if it is indeed related to the gap—would have to be reinterpreted as a 4Δ absorption.

In the two-component picture, nearly all the free-carrier oscillator strength appears under the zero-frequency delta function. Finite frequency absorption is due to the midinfrared band. In this picture, the shoulder which develops in $\mathcal{T}_s$ around 700 cm^{-1} is entirely due to the midinfrared oscillator and the collapse of the Drude peak to a delta function. The peak in $\mathcal{T}_s$ represents a passband between absorbing regions at zero and midinfrared frequencies. No spectroscopic gap is discernible in the infrared transmittance.

Accordingly, the observed feature around 700 cm^{1} can be interpreted as a consequence either of a two-component dielectric function or of 4Δ excitations from the superconducting condensate. For the latter interpretation, any gap in the optical conductivity has to be relatively smooth and independent of temperature, contrary to simple BCS behavior. In that sense, the two models are very similar: some kind of bound (either optically active or optically inactive) excitation, with binding energy of the order of 1000 K, exists well above T_c.

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