Far-infrared transmission of $Bi_2Sr_2CaCu_2O_8$ films

R. A. Hughes and T. Timusk
Institute for Materials Research, McMaster University, Hamilton, Ontario, Canada L8S 4M1

S. L. Cooper, G. A. Thomas, J. J. Yeh, and M. Hong
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
(Received 24 April 1989)

The far-infrared transmission spectra for high-conductivity $Bi_2Sr_2CaCu_2O_8$ films with the radiation polarized in the *ab* plane have been measured for temperatures above and below T_c. The normal-state properties show a Drude-like response. The transmission spectrum can be used to derive both the dc conductivity (90 $\mu\Omega$ cm at T_c) and the relaxation rate (165 cm^{-1} at T_c). If an energy gap at $3.5kT_c$ or higher is assumed then the film is in the clean limit. In the superconducting state the transmission drops at low frequency but remains finite.

Far-infrared transmission experiments on thin films are a well-established and informative method of probing superconducting materials.[1-3] Up to now, the optical work on the high-T_c compounds has been dominated by reflectance measurements, while transmission experiments have remained an essentially unexplored field due to the stringent restrictions imposed on the samples. The films have to be thin, of the order of a few hundred Å in thickness, and deposited upon substrates that are transparent to infrared radiation but at the same time suitable for epitaxial growth. Such films deposited on MgO have recently become available.[4] The work presented here shows the temperature-dependent transmission spectra for sputtered films of the $Bi_2Sr_2CaCu_2O_8$ compound.

200-Å thick $Bi_2Sr_2CaCu_2O_8$ films were deposited on 0.5 mm thick MgO by rf magnetron sputtering.[4] The films were oriented with their *c*-axes perpendicular to the underlying substrate. They exhibited an onset temperature of 85 K with a transition width of 5 K. The samples were mounted on a brass shim over a hole, 2–4 mm in diameter. The radiation was allowed to pass through the superconducting film and then through the substrate. The MgO substrate introduces complications as well as limitations to the experiment. First of all, interference effects between light reflected off the front and back surfaces of the MgO layer are clearly visible. The removal of these effects limits the spectral resolution. The reststrahlen band of MgO is at 350 cm^{-1} and restricts measurements to frequencies below 300 cm^{-1}. In addition, the substrate properties are slightly temperature dependent. Most of these effects can be removed by referencing the sample spectrum to an identical MgO substrate. It should be emphasized that the spectra obtained in this manner are qualitatively similar, but not equivalent to that of a free-standing $Bi_2Sr_2CaCu_2O_8$ film due to the $Bi_2Sr_2CaCu_2O_8$/MgO interface. To obtain quantitative information from these data the optical properties of the substrate must also be measured. To this end, a temperature-dependent transmission experiment was performed on the substrate material by referencing it to a circular aperture. The real and imaginary parts of the index of refracton of MgO derived from these data showed good agreement with those of Jasperse *et al.*[5]

Figure 1 shows the transmission spectra of two $Bi_2Sr_2CaCu_2O_8$ films (samples *A* and *B*) with the radiation polarized in the *ab* plane for temperatures above and below the superconducting transition. The spectra have been referenced to a MgO substrate. The transmission increases monotonically with frequency. The onset of superconductivity is characterized by a sharp drop below T_c in transmittance at low frequencies and a less dramatic rise in the transmittance at higher frequencies. The crossover where the normal and superconducting states have equal transmission occurs near 180 cm^{-1}. Both samples show very little temperature dependence below 60 K. Samples *A* and *B* show behavior which is qualitatively similar, but sample *B* is far more transmissive. We measured several additional samples all of which showed qualitatively similar behavior.

None of the samples have the behavior expected for a conventional superconductor: zero transmission when extrapolated to zero frequency. It is felt that this discrepancy arises from the presence of nonsuperconducting material within the sample. It is not clear what the form of the nonsuperconducting portion is but one possibility, consistent with the nonzero transmission observed in both samples, is finite areas, larger than the wavelength, of insulating or poorly conducting metallic material that would provide a parallel path for the radiation. The fact that sample *B* is more transmissive is an indication that it is of lower quality. This nonideal behavior is also observed in microwave-loss experiments on high-T_c materials at low temperature where the loss exceeds what is expected from BCS theory by several orders of magnitude. We found that this residual transmission in the superconducting state was highly sample dependent and could often be reduced by an oxygen anneal.

In order to extract quantitative information from the spectra, substrate effects must be dealt with in a more rigorous manner. The problem can be described in terms of a frequency-dependent index of refraction ($N=n-ik$) describing both the film and its underlying substrate, here subscript 0 refers to vacuum, 1 to the substrate, and 2 to the film. In this case, the transmission through the two

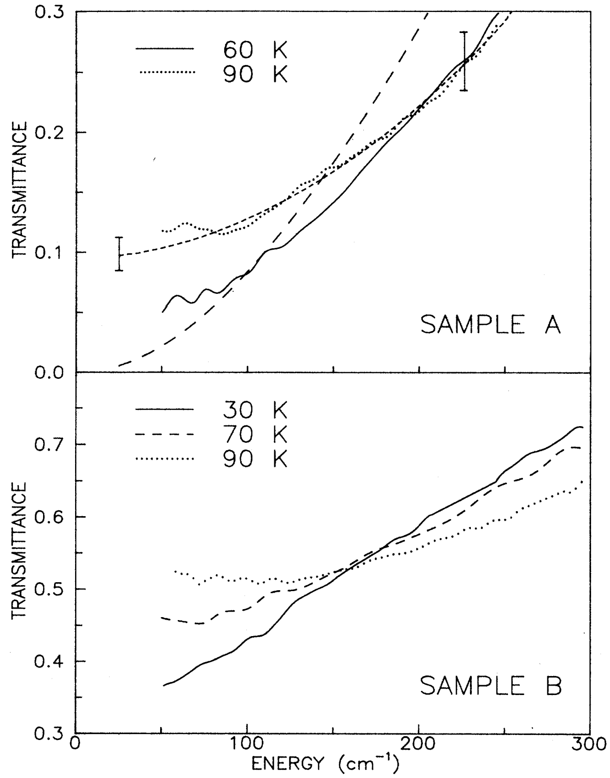

FIG. 1. Far-infrared transmittance of two samples of 200-Å $Bi_2Sr_2CaCu_2O_8$ films for temperatures above and below T_c. Top panel shows sample A's experimental transmission and a fit with a Drude model (short-dashed curve). The error bars shown demonstrate the effect of varying the dc conductivity by $\pm 10\%$ (low-frequency error bar), and the relaxation rate by $\pm 10\%$ (high-frequency error bar). The long-dashed curve is a calculated transmission curve for a BCS superconductor in the clean limit. Sample B, lower panel, is far more transmissive than sample A over all frequencies.

layers is given by[6]

$$T = \frac{n_0}{n_3} |t|^2 , \quad (1)$$

where

$$t = \frac{t_{32} t_{21} t_{10} \exp[-i(\beta_1 + \beta_2)]}{(1 + r_{32} r_{21} e^{-i2\beta_2}) + (r_{21} + r_{32} e^{-i2\beta_2}) r_{10} e^{-i2\beta_1}} ,$$

$$r_{ij} = \frac{N_i - N_j}{N_1 + N_j}, \quad t_{ij} = \frac{2N_i}{N_i + N_j}, \quad \text{and } \beta_k = \frac{2\pi}{\lambda} N_k d_k .$$

Here λ describes the wavelength of the incident radiation in vacuum and d_k is the thickness of layer k. The values for the optical constants n_1 and k_1 were determined in an independent experiment on an identical MgO substrate. Thus, the only unknown variables in Eq. (1) are the optical constants for the $Bi_2Sr_2CaCu_2O_8$ film. Fits to the data can be performed using a model for N_2, the optical response of the film, and comparing the transmission calculated from Eq. (1) with the experiments.

Reflectance measurements on single-crystal $Bi_2Sr_2CaCu_2O_8$ (Ref. 7) have shown that its infrared conductivity can be described with a two-component model: a low-frequency temperature-dependent Drude portion and a temperature-independent mid-infrared band that can be described by broad harmonic oscillators. A similar model has been used for single-crystal $YBa_2Cu_3O_{7-\delta}$.[8,9] The spectral region investigated in this research corresponds to the regime where the oscillators play a minor role and a simple Drude model, where $4\pi\sigma_1 = \omega_p^2/[\Gamma(1+\omega^2/\Gamma^2)]$, gives a satisfactory description.

A fit to the 90-K data for sample A (shown as a short-dashed curve in Fig. 1) gives a plasma frequency (ω_p) of 10 500 cm^{-1} and a relaxation rate (Γ) of 165 cm^{-1}. From these parameters, using the Drude formula $4\pi\sigma_0 = \omega_p^2/\Gamma$, the dc resistivity is found to be 90 $\mu\Omega$ cm. These results are in good agreement with the dc resistivity measurements of $Bi_2Sr_2CaCu_2O_8$ films[4] and crystals[10] as well as optical measurements on $Bi_2Sr_2CaCu_2O_8$ crystals.[7] Reflectance measurements on single crystals gave $\omega_p = 11\,600$ cm^{-1} and $\Gamma = 69$ cm^{-1}.

Since the thin-film transmission experiments provide a straightforward method for finding the plasma frequency and the relaxation rate it is of interest to study the temperature dependence of these quantities in the normal state. We found, in general, the plasma frequency to be temperature independent for these films. The relaxation rate was also relatively temperature independent except in one sample where a linear dependence was observed similar to what has been seen in ceramic samples.[8] The dc conductivity, measured with a four-probe technique, in general showed a linear temperature dependence. Thus, in contrast to some Hall-effect measurements where a temperature-dependent carrier density can be inferred from the temperature-dependent Hall constant, all the evidence from the far-infrared response in the normal state points to a temperature-independent carrier plasma frequency and carrier concentration.

Theoretical fits to the superconducting state are more difficult. First of all, a definitive theory describing the oxide superconductors has not emerged. Second, a simple analysis in terms of the BCS formalism is not valid for these films due to the regions of normal material responsible for the lack of true BCS response which is characterized by a zero transmission at zero frequency. The long-dashed curve in the top panel of Fig. 1 shows the expected transmission for a BCS superconductor with a normal-state relaxation rate of 10 cm^{-1}. The fit to the experimental 60-K curve is poor and the task of extracting a gap from the measurements is very dubious.

In conventional superconductors, where a division of the superconducting-transmission spectrum (T_S) by the normal-state spectrum (T_N) gives rise to a peak at the energy-gap frequency, transmission experiments provide a straightforward method of determining the energy gap. While the ratio T_S/T_N for the $Bi_2Sr_2CaCu_2O_8$ compound as derived from Fig. 1 does rise from a low value to a value in excess of one, as in conventional superconductors, the exact behavior of this ratio beyond 300 cm^{-1} is unknown since the substrate becomes opaque, but it is expected that it will approach one in the high-frequency limit. Thus, a peak must appear in the ratio T_S/T_N and could be interpreted as an energy gap in excess of 300 cm^{-1} or $2\Delta/kT_c > 5$. However, we feel that this method of determining the energy gap has severe shortcomings due to the fundamental differences between the high-T_c materials and their low-T_c counterparts.

These differences lead to several complications. First, the normal-state properties are not known at low temperatures since a magnetic field cannot be used to destroy the superconductivity in the high-T_c compounds due to the high-critical fields. As a result the ratio T_S/T_N, where T_S and T_N are measured at different temperatures, can show structure which is solely due to the variation of normal-state properties with temperature. Thus, any gap derived from the ratio T_S/T_N should be met with some skepticism. A second complication arises from the fact that the relaxation rate of 165 cm^{-1} derived from the Drude fit to the normal state may place the material in the clean limit ($\Gamma < 2\Delta$) as is the case if the gap size is $3.5kT_c$ or larger. In this limit the gap feature in the conductivity tends to become unobservable by ordinary optical techniques.

Figure 2 shows the results of model calculations which demonstrate the difficulty of identifying a gap in the clean limit. The optical conductivity used to calculate T_N was assumed to be Drude-like with a plasma frequency of 10 500 cm^{-1} and a relaxation rate of 165 cm^{-1}. In addition, the oscillators needed to describe the high-frequency response were included in this calculation. The parameters used to describe these oscillators were taken from the work of Reedyk *et al.*[7] T_S was calculated using the Leplae formula[11] describing the BCS response with an energy gap of $2\Delta = 3.5kT_c = 210$ cm^{-1}. Below T_c it is expected that the normal-state relaxation rate will decline in concert with the linear drop in the resistivity. In order to simulate this effect the superconducting-state properties were derived for a series of relaxation rates below the normal-state value. The calculated curves show the same qualitative features as the spectra of samples A and B in that the superconducting transmittance rises more rapidly than the normal state and there is a crossover in the 100–200-cm^{-1} region. It is clear, however, that while in the dirty limit $\Gamma \gg 2\Delta$, a distinct feature in the transmission ratio can be seen at the gap frequency; this feature weakens as the clean limit of $\Gamma \ll 2\Delta$ is approached. A false peak appears above the gap frequency. Its position depends on the relative strength of the mid-infrared band and the Drude absorption.

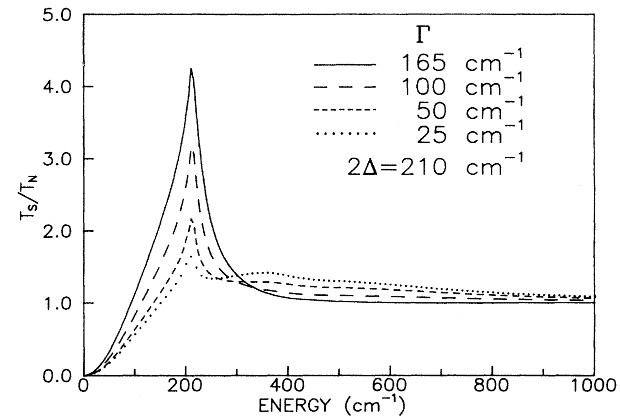

FIG. 2. The ratio of superconducting to normal transmittance for a superconductor in the clean limit ($\Gamma < 2\Delta$). The energy gap was chosen to be 210 cm^{-1}. Note that the energy-gap feature disappears in the very clean limit.

Another potential problem comes from the comparison of spectra at two different temperatures. It is easy to see that even the ratio of two normal metal transmittance curves with different relaxation rates will mimic a T_S/T_N ratio. The fact that the superconducting transmittance rises above the normal-state values in the clean limit (the ratio rises above unity) is not an energy-gap feature, but instead arises from the fact that two normal-state relaxation rates are involved in the problem. If the temperature-dependent relaxation rate is not taken into account this feature could be falsely labeled as an energy gap.

In conclusion, far-infrared transmission spectra for $Bi_2Sr_2CaCu_2O_8$ films show a Drude response in the normal state for frequencies below 300 cm^{-1}. The transmittance drops in the superconducting state at low frequencies and gradually rises above the normal-state value as frequency is increased. There is a residual absorption in the superconducting state which seems to be a sample-dependent factor, possibly due to a chemical instability in the very thin films used for these measurements.

[1] R. E. Glover III and M. Tinkham, Phys. Rev. **104**, 844 (1956).
[2] M. Tinkham, Phys. Rev. **104**, 845 (1956).
[3] R. E. Glover III and M. Tinkham, Phys. Rev. **108**, 243 (1957).
[4] J. J. Yeh and M. Hong, Appl. Phys. Lett. **54**, 769 (1989).
[5] J. R. Jasperse, A. Kahan, J. N. Plendl, and S. S. Mitra, Phys. Rev. **146**, 526 (1956).
[6] L. Ley, in *Topics in Applied Physics*, edited by J. D. Joannopoulos and G. Lucovsky (Springer-Verlag, New York, 1984), p. 61.
[7] M. Reedyk, D. A. Bonn, J. D. Garett, J. E. Greedan, C. V. Stager, T. Timusk, K. Kamarás, and D. B. Tanner, Phys. Rev. B **38**, 11 981 (1988).
[8] T. Timusk, S. L. Herr, C. D. Porter, D. B. Tanner, D. A. Bonn, J. D. Garrett, J. E. Greedan, and M. Reedyk, Phys. Rev. B **38**, 6683 (1988).
[9] G. A. Thomas, J. Orenstein, D. H. Rapkine, M. Capizza, A. J. Millis, R. N. Bhatt, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. **61**, 1313 (1988).
[10] S. Martin, A. T. Fiory, R. M. Fleming, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. **60**, 2194 (1988).
[11] Luc Leplae, Phys. Rev. B **27**, 1911 (1983); R. A. Klemm, K. Scharnberg, D. Walker, and C. T. Rieck, Z. Phys. B **72**, 139 (1988).