PHYSICAL REVIEW B VOLUME 37, NUMBER 4 1 FEBRUARY 1988

Temperature-dependent far-infrared reflectance of La-Sr-Cu-O and La-Ca-Cu-O: Bardeen-Cooper-Schrieffer electrodynamics but uncertain energy gap

M. S. Sherwin, P. L. Richards, and A. Zettl
Department of Physics, University of California, Berkeley, California 94720
and Materials and Chemical Sciences Division, Lawrence Berkeley Laboratory, Berkeley, California 94720
(Received 21 September 1987)

The available far-infrared data for polycrystalline La-Sr-Cu-O and La-Ca-Cu-O show a reflectance edge with energy near $2.5k_BT_c$. This edge has been variously interpreted as the onset of absorption due to an energy gap, and as a low-frequency plasma edge caused by strong far-infrared resonances. Our measured temperature dependence of the reflectance edge closely fits the temperature dependence of the order parameter in a mean-field theory, and hence is consistent with the energy-gap hypothesis. In this paper, we construct a model dielectric function for $La_{1.85}Sr_{0.15}CuO_4$ which is consistent with mean-field theory and the hypothesis of a plasma edge. We find that the temperature dependence of the plasma frequency in this model also closely fits the measured temperature dependence of the reflectance edge. Furthermore, both hypotheses accurately predict the experimentally observed temperature dependence of the absorption at frequencies much less than the reflectance edge. This observation has significant implications for the construction of fast low-loss superconducting devices. We conclude that the electrodynamics of the superconducting transition in $La_{1.85}Sr_{0.15}CuO_4$ are well described by a Bardeen-Cooper-Schrieffer-like mean-field theory. However, given the identical predictions of the energy-gap and plasma-edge hypotheses, it is premature to deduce a precise value for the magnitude of the energy gap from the infrared data.

INTRODUCTION

A vigorous research effort into the electrodynamics of high-T_c superconductors has been fueled by the great scientific and technological importance of this subject. The existence and magnitude of the superconducting energy gap are crucial scientific issues that have traditionally been illuminated by far-infrared (FIR) spectroscopy. The possibility of devices that operate with low resistive loss in the (100–1000)-GHz frequency range at liquid-nitrogen temperature is of technological importance. In this paper we use experimental and theoretical results on the temperature dependence of the FIR reflectance of polycrystalline $La_{1.85}(Sr,Ca)_{0.15}CuO_4$ to address the following questions. First, can one extract an energy gap from the FIR reflectance of high-T_c superconductors? Second, how well do the temperature-dependent electrodynamics of the BCS theory fit high-T_c superconductors? Third, do the ac losses for frequencies much less than the energy gap predicted by BCS scale with temperature as predicted by BCS?

Although the mechanism for superconductivity in high-T_c superconductors is not yet clear, experiments have provided important guidelines. Measurements on Josephson junctions[1] indicate that current is carried by pairs of electrons. However, the absence of an observable isotope shift[2] in T_c in $YBa_2Cu_3O_7$ and the small size of the oxygen isotope shift[3] in $La_{1.85}Sr_{0.15}CuO_4$ indicates that phonons alone probably do not mediate the pairing. It has been suggested that a BCS-like theory in which excitons or other relatively-high-energy excitations mediate electron pairing may apply.[4] The electrodynamics predicted by this class of theories should be close to that of BCS theory. In particular, a mean-field treatment of these theories should predict an energy gap with a temperature dependence and magnitude identical to that predicted by BCS. The magnitudes of the energy gap extracted by tunneling for[5] $La_{1.85}Sr_{0.15}Cu_4$ and[6] $Y_1Ba_2Cu_3O_7$ have been 4.5–9 and 3.7–5.6, respectively, consistent with a strong-coupling pairing theory. In contrast, Anderson[7] has suggested that there may be no observable energy gap in high-T_c superconductors if they are described by a resonating-valence-bond model.

We begin with a review of relevant and sometimes conflicting interpretations of the FIR reflectance of $La_{1.85}Sr_{0.15}CuO_4$. Many groups[8-11] have observed a reflectance edge near 50 cm^{-1} in the superconducting state of $La_{1.85}Sr_{0.15}CuO_4$. This edge was first assigned to the onset of absorption due to excitations across a superconducting energy gap. The magnitude of the energy gap extracted by the first such assignments[8-10] was from $1.6k_BT_c$ to $2.7k_BT_c$, considerably smaller[11,12] than the BCS prediction of $3.5k_BT_c$ and the tunneling measurements.[5] Recently, an entirely different mechanism for the 50-cm^{-1} edge in $La_{1.85}Sr_{0.15}CuO_4$ has been proposed by Bonn *et al.*[13] They have assigned this edge to a zero crossing of the real part of the dielectric function ϵ_1, caused by a strong resonance at 240 cm^{-1} and a weaker resonance at 500 cm^{-1}. Under this interpretation, neither the existence nor the value of the energy gap are obvious from far-infrared reflectance data.

A priori, one might think that the temperature dependence of the 50-cm^{-1} reflectance edge could be used to distinguish between the hypothesis of an energy gap and the hypothesis of a plasmon. We report here measurements of the FIR reflectance of polycrystalline samples of

$La_{1.85}Sr_{0.15}CuO_4$ and $La_{1.85}Ca_{0.15}CuO_4$. We have probed the temperature dependence of the FIR reflectance of $La_{1.85}Sr_{01.5}CuO_4$. At temperatures well below T_c, we find a reflectance edge whose frequency scales with T_c in different materials. The frequency of this edge in a given material scales with temperature as the BCS gap. We also describe a model to determine the temperature dependence of the reflectance edge predicted by the plasmon hypothesis. The superconducting state is modeled using the temperature-dependent theory of Mattis and Bardeen, which should hold for any mean-field pairing theory of superconductivity, independent of the nature of the coupling. We find that the plasmon and the energy-gap hypotheses give nearly identical predictions for the temperature dependence of the frequency of the reflectance edge. The predictions for the temperature dependence for both theories agree with experimental results. We conclude that a mean-field, BCS-like theory of the electrodynamics of high-T_c superconductors is consistent with the FIR data. However, it seems premature to deduce an energy gap from FIR data.

The most technologically significant result of this paper is that our experimentally observed temperature dependence of the absorption in the superconducting state of polycrystalline $La_{1.95}Sr_{0.15}CuO_4$ for frequencies well below the reflectance edge seems well described by the temperature-dependent Mattis-Bardeen theory, or equivalently by a two-fluid model. If this result holds for $Y_1Ba_2Cu_3O_7$, it imposes constraints on the operating temperature of fast superconducting devices made from this material.

EXPERIMENT

The samples used for the measurements reported here are 1-cm-diam ceramic pellets of $La_{1.85}Sr_{0.15}CuO_4$ and $La_{1.85}Ca_{0.15}CuO_4$. At low temperatures the Sr-doped sample showed a large volume exclusion of a magnetic field, indicating that a large fraction of the sample consisted of a superconducting phase. The Ca-doped sample excluded only $\frac{1}{5}$ as much of the magnetic field as the Sr-doped sample. The details of the sample preparation have been described elsewhere.[8] The Sr- and Ca-doped samples showed the onset of superconductivity at $T_{co}=36$ and 17 K, respectively, as determined from magnetic-susceptibility (χ) measurements. The superconducting phase transition was also observed directly from the FIR measurements in the Sr-doped sample. The reflectance near 15 cm^{-1} began to increase sharply as a function of decreasing temperature at 37 ± 1 K (see Fig. 5). This temperature is in agreement with the value of T_{co} determined from $\chi(T)$, and thus we scale all temperature- and material-dependent properties to T_{co}.

FIR reflectance measurements were performed with a Michelson interferometer, adapted to a helium-gas-flow cryostat to allow sample-temperature variations from 6 K to room temperature. During the experiment, chopped radiation with a 10° angle of incidence was detected after a single reflection off the sample surface by a sensitive composite bolometer operated at 1.2 K. At each sample temperature for which a reflectance spectrum was recorded the data were normalized to a polished brass mirror. After all measurements on $La_{1.85}Sr_{0.15}CuO_4$ had been performed, we attempted to correct our results for the effects of surface scattering by evaporating metal onto the sample surface and using the metallized sample as a reference. The reflectance of the metallized sample was independent of frequency for frequencies less than 60 cm^{-1}, indicating that our data in this frequency range are not much affected by the roughness of the sample surface. Above 60 cm^{-1}, however, the reflectance of the metallized sample decreased continuously with increasing frequency, indicating that our absolute reflectance data in this frequency range are significantly affected by surface scattering. Small cracks occurred in the sample surface just before metallization, which prevented an accurate final normalization. However, the conclusions of this paper are not affected by surface scattering: We are investigating changes in the reflectance as a function of temperature, and thus we are relatively insensitive to temperature-independent losses due to sample geometry.

Figure 1(a) shows the ratio R_s/R_n of the superconducting to the normal-state reflectance for $La_{1.85}Sr_{0.15}CuO_4$. Here, R_s and R_n are the reflectances measured at 6 and 52 K, respectively, well below and well above T_{co}. R_s/R_n is greater than 1 for frequencies less than 60 cm^{-1} [$=2.4k_BT_{co}$(Sr)]. As the frequency increases past 60 cm^{-1}, R_s/R_n drops below unity, reaches a minimum at 70 cm^{-1}, and then approaches unity from below. This behavior is consistent with that reported by many groups.[9-11,13] Figure 1(b) shows the first published reflectance data for $La_{1.85}Ca_{0.15}CuO_4$. We have expanded the frequency scale of Fig. 1(b) relative to 1(a) by the ratio of the transition temperatures of the two materials $T_{co}(Sr)/T_{co}(Ca)=36/17$. The behavior of R_s/R_n is similar to that for the Sr-doped material, with characteristic frequencies scaled by T_{co}. Below 30 cm^{-1}, R_s/R_n is greater than 1. Between 30 cm^{-1} [$=2.4k_BT_{co}$(Ca)] and 40 cm^{-1} [$=3.2k_BT_{co}$(Ca)], R_s/R_n drops below unity. The deviations of R_s/R_n from unity are smaller for the Ca- than for the Sr-doped sample. These differences may arise from the fact that, based on magnetic measurements, a smaller fraction of the Ca-doped sample was of a superconducting phase.

Figure 2 shows a series of normalized reflectance spectra of $La_{1.85}Sr_{0.15}CuO_4$ for frequencies 10–90 cm^{-1} at selected temperatures above and below $T_{co}=36$ K. In the low-temperature regimes well below T_{co}, the reflectance follows a consistent behavior. At low frequencies, R is near unity and decreases only slightly with increasing frequency. At higher frequencies, R drops sharply at a characteristic frequency f_0 and begins to flatten out once again at an even higher characteristic frequency f_1. At 6 K, f_0 and f_1 are clearly identified at 50 and 66 cm^{-1}, respectively. Both f_0 and f_1 decrease with increasing temperature above 6 K. Above 36 K, f_0 and f_1 are no longer clearly identifiable. At 52 K, the reflectance R decreases smoothly with increasing frequency. Above 50 K, the reflectance curve was found to be rather insensitive to temperature.

If one assumes (as is done in Refs. 8–12) that the real part of the dielectric function is negative throughout the

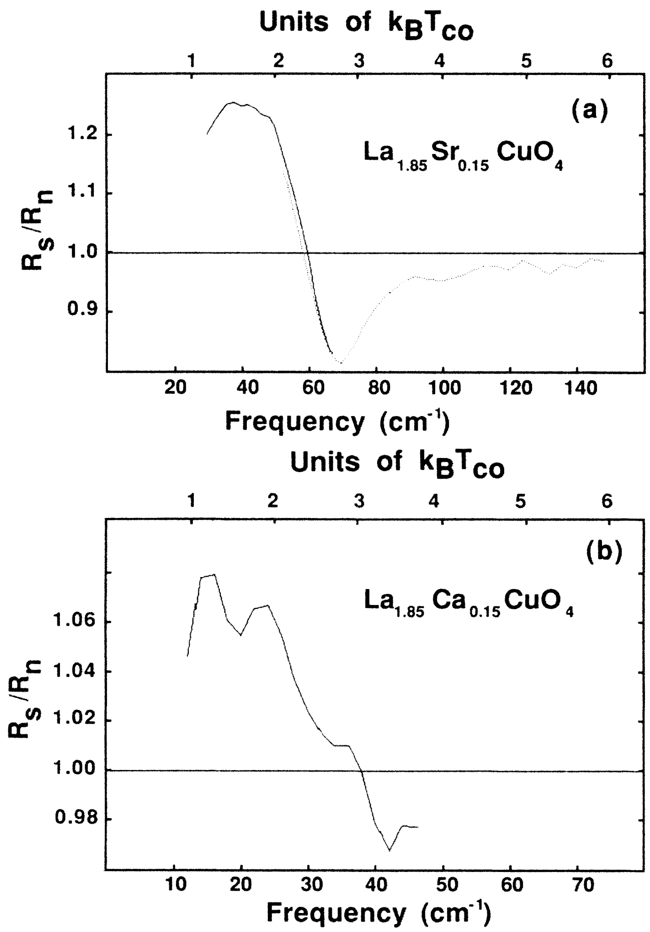

FIG. 1. The ratio R_s/R_n of the superconducting to normal-state reflectance as a function of frequency for $La_{1.85}Sr_{0.15}CuO_4$ and $La_{1.85}Ca_{0.15}CuO_4$. A solid line has been drawn to mark $R_s/R_n=1$. The scale of (b) is expanded relative to that of (a) by the ratio of the superconducting onset temperatures $T_{co}(Sr)/T_{co}(Ca)=(36\text{ K})/(17\text{ K})$, showing that the frequency of the characteristic features of R_s/R_n scales with T_{co}. (a) $La_{1.85}Sr_{0.15}CuO_4$: R_s and R_n are reflectances measured at 5 and 52 K. The solid and dotted lines represent data from different experimental runs. (b) $La_{1.85}Ca_{0.15}CuO_4$: R_s and R_n are reflectances measured at 9 and 24 K, respectively.

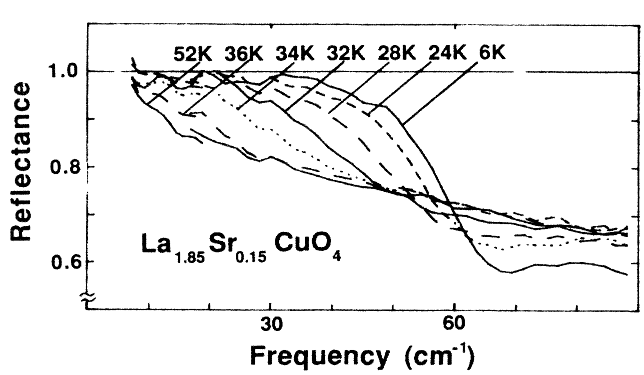

FIG. 2. Reflectance of $La_{1.85}Sr_{0.15}CuO_4$ at selected temperatures above and below T_{co}. The temperature-dependent reflectance edge has been interpreted as an energy gap (Refs. 8–12) and as a plasma edge (Ref. 13). A solid line marks $R=1$.

FIR, then a general interpretation of the spectra in Figs. 1 and 2 is straightforward. The reflectance feature windowed by f_0 and f_1 can be identified as the onset of photon absorption at the superconducting energy gap 2Δ. A similar interpretation is possible for the $La_{1.85}Ca_{0.15}CuO_4$ data shown in Fig. 1(b). The fact that the frequency of these features in the two materials at temperatures much less than T_{co} scales with T_{co} is consistent with the energy-gap hypothesis. As we shall see below, the temperature dependence of the reflectance edge is also consistent with the hypothesis of a BCS-like energy gap.

MODEL FOR THE REFLECTANCE EDGE

In the remainder of this paper we construct a model to investigate the temperature dependence of the reflectance predicted by the plasmon hypothesis, and we compare its predictions to those of the energy-gap hypothesis. We note that the temperature dependence of the plasmon hypothesis has not been investigated previously. Finally, we investigate the scaling of the reflectance of $La_{1.85}Sr_{0.15}CuO_4$ near 16 cm^{-1} (500 GHz) with temperature for both the energy-gap and plasmon hypotheses.

The interpretation of the normal-state reflectance of polycrystalline samples of $La_{1.85}Sr_{0.15}CuO_4$ is complicated by the fact that the crystal structure of this material is highly anisotropic. The conduction electrons are thought to be mostly confined to sheets parallel to the *a*-*b* plane, with a relatively low conductivity perpendicular to the *a*-*b* plane. Two approaches have been taken to account for the effect of anisotropy on FIR spectra of polycrystalline samples, and these approaches yield different assignments for the observed features in the reflectance spectra. Thomas *et al.*[14] have argued that since the size of typical crystallites is much smaller than a wavelength at FIR frequencies, a long-wavelength effective-medium theory should apply. In such a theory, the reflectance is calculated from a dielectric function which is an average over all crystalline orientations. Under this interpretation, the 240-cm^{-1} resonance first reported by Bonn *et al.* must have an extremely large oscillator strength and must have components both in the *a*-*b* plane and perpendicular to the *a*-*b* plane.[15] Furthermore, if one interprets the 50-cm^{-1} edge in the superconducting state as a plasma edge, this feature must also have components both in and out of the *a*-*b* plane. Schlesinger *et al.*[16] have analyzed the spectra of their polycrystalline samples by using a short-wavelength approximation and averaging reflectivities over different crystallite orientations, rather than averaging the dielectric function. This approach is combined with the assumption that the normal-state reflectance of $La_{1.85}Sr_{0.15}CuO_4$ should closely resemble that of La_2NiO_4, a material of the same crystal structure on which FIR measurements of a single crystal have been made.[17] In La_2NiO_4, there is a moderate oscillator strength resonance near 250 cm^{-1} which has components *only* perpendicular to the *a*-*b* plane. Schlesinger assigns the 240-cm^{-1} resonance to vibrations perpendicular to the *a*-*b* plane, and assigns the temperature-dependent reflectance edge near 50 cm^{-1} to a plasma oscillation also perpendicular to the a-b plane. The first infrared data on single crystals and oriented films of the related Y-Ba-Cu-O compound show that the phonon peaks in that case are confined to the c axis.[18]

The analyses of Thomas[15] and Schlesinger *et al.*[16] both assume a low-frequency plasmon. The differences are that the two analyses give different assignments to the direction of the plasma oscillation, and different oscillator strengths for the 240-cm^{-1} mode are needed to fit the normal-state spectra. In our modeling, since we are concerned with wavelengths larger than 50 μm (much greater than the size of the <10-μm crystallites), we adopt a long-wavelength approximation in which the reflectance is given by the standard formula $R = |(\epsilon^{1/2}-1)/(\epsilon^{1/2}+1)|^2$, where R is the reflectance and $\epsilon = \epsilon_1 + i\epsilon_2$ is an average dielectric function.

In order to convincingly model the reflectance in the normal state, it is necessary to consider the reflectance over a broad frequency range. We have chosen the best available data in each frequency range. We use our data in the (10–90)-cm^{-1} range, those of Bonn *et al.*[13] (which are close to our reported data from 50 to 90 cm^{-1}) from 90 to 1000 cm^{-1}, and those of Orenstein *et al.*[19] from 1000 to 24 000 cm^{-1}. The most important conclusions of our modeling will prove to be insensitive to the details of the data above 1000 cm^{-1}.

In the normal state, we model the reflectance for frequencies less than 200 cm^{-1} with a Drude term for the free carriers, a Lorentz oscillator for the 240-cm^{-1} mode deduced by Bonn *et al.*, and a background dielectric constant for the oscillator strength at much higher frequencies than the FIR. Thus,

$$\epsilon_1(\omega) = \frac{-\omega_p^2\tau^2}{1+\omega^2\tau^2} + \frac{\omega_T^2(\omega_1^2-\omega^2)}{(\omega_1^2-\omega^2)+\omega^2\tau_1^2} + \epsilon_\infty , \qquad (1)$$

$$\epsilon_2(\omega) = \frac{\omega_p^2\tau}{\omega(1+\omega^2\tau^2)} + \frac{\omega_t^2(\omega/\tau_1)}{(\omega_1^2-\omega^2)+\omega^2/\tau_1^2} , \qquad (2)$$

where ϵ_1 and ϵ_2 are the real and imaginary parts of the dielectric function, ω is the incident-photon frequency, ω_p and τ are the plasma frequency and relaxation time of the free carriers, ω_T, ω_1, and τ_1 are the plasma frequency, resonant frequency, and relaxation time of the Lorentz oscillator, and ϵ_∞ is the background dielectric constant.

We determine the parameters of our normal-state fit as follows. Orenstein *et al.* fitted their high-frequency reflectance data to a model in which 94% of the oscillator strength of the electrons in the conduction band is associated with a (nonsuperconducting) gap at 3600 cm^{-1} which contributes a constant $\epsilon_1 = 20$ in the FIR. The remaining 6% is associated with a term of the Drude form. We have parametrized the FIR resonances at 240 and 500 cm^{-1} deduced by Bonn *et al.* by Lorentz oscillators and have added them to the model of Orenstein *et al.* We adjusted the oscillator strength of the Drude term so that the sum of the oscillator strengths of the Drude term and the two resonances adds up to 6% of the total oscillator strength in the conduction band proposed by Orenstein *et al.* In the (0–100)-cm^{-1} range the resonance at 500 cm^{-1} can be modeled by a constant contribution to ϵ_1 of 3.

The parameters of our model are as follows: $\omega_p = 3350$ cm^{-1}, $\omega_T = 1860$ cm^{-1}, $\omega_1 = 239$ cm^{-1}, $1/\tau_1 = 33$ cm^{-1}, and $\epsilon_\infty = 23$. The relaxation time τ of the free carriers is adjusted to fit our normal-state data. We find that $1/\tau = 2000$ cm^{-1} gives an adequate fit to our data and those of Bonn *et al.*[13] in the frequency range 10–100 cm^{-1}. Given the inhomogeneous nature of these samples, a detailed fit to the reflectance is not warranted. We note that our model predicts a *positive* ϵ_1 at zero frequency in the normal state: the ability of heavily damped free carriers to screen electromagnetic radiation is overwhelmed by the polarizability of the FIR resonances.

Once we fit the normal-state reflectance, we calculate the reflectance at selected temperatures in the superconducting state with no additional adjustable parameters. To model the reflectance in the superconducting state, we assume an energy gap of magnitude and temperature dependence predicted by weak-coupling BCS [i.e., $2\Delta(T=0) = 3.5k_BT_{co}$]. The Drude terms in Eqs. (1) and (2) are replaced with terms calculated from the temperature-dependent Mattis-Bardeen[20] expressions for the frequency-dependent conductivity in the superconducting state. We have integrated numerically these singular integrals using Gaussian and Chebyshev integration routines.[21] The Mattis-Bardeen expressions are valid in the limits in which the penetration depth of the electromagnetic radiation is much larger than or much less than the coherence length. The former limit applies in the superconducting oxides.

The temperature-dependent reflectivity from our plasmon model is shown in Fig. 3. Many qualitative features of the data are apparent. Below $T = T_c$, the calculated reflectance shows systematic trends similar to the data of Fig. 2. For low frequencies, the reflectance is near unity. At a frequency f_0 the reflectance begins to drop and at f_1 it begins to flatten out. For $T < 34$ K there is a

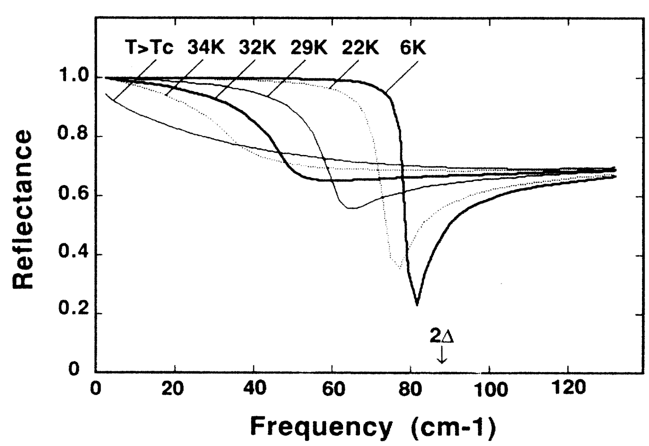

FIG. 3. Reflectance calculated at selected temperatures from a model which contains free carriers and a strong phonon, and treats the reflectance edge as a plasma edge. The model qualitatively reproduces the experimental data of Fig. 2. The position of the energy gap $2\Delta(T=0)$ used in this calculation is marked with an arrow. There is no obvious feature in the calculated reflectance at this frequency.

minimum. The reflectance then approaches the normal-state reflectance from below. The steepness of the reflectance edge increases with decreasing temperature. At $T=6$ K the reflectance edge at 80 cm^{-1} is extremely sharp. The experimentally observed reflectance edge is broader, which is to be expected if sample inhomogeneity leads to damping mechanisms not included in the model. The assumed energy gap for $T=6$ K is 88 cm^{-1} and is marked by an arrow in Fig. 3. There is no obvious feature in the calculated reflectance at this frequency. This shows that if the plasmon hypothesis is correct, it is impossible to extract a value for the energy gap by simple inspection of the reflectance.

The derivatives of the reflectance curves in Figs. 2 and 3 show clear minima at a frequency f_p between f_0 and f_1 and thus enable an objective comparison of the temperature dependence of the reflectance edge in the model and the experiment. We have plotted the temperature dependence of f_p for both experiment and model in Fig. 4. We have also plotted the temperature dependence of the BCS gap. All quantities are normalized to 1 for temperatures much less than T_{co}. We see that the temperature dependence of the reflectance edge in both the model and the experiment closely fits the temperature dependence of the BCS gap. Thus the predicted temperature dependencies of the reflectance edge in the plasmon hypothesis and the energy-gap hypothesis are *virtually indistinguishable.* The only difference between the temperature-dependence predictions of the two hypotheses is that f_p in the plasmon hypothesis lies at slightly higher frequencies than the BCS curve for temperatures $T_{co}/3<T<T_{co}$. This arises in the model from the frequency dependence of the contribution to ϵ_1 of the 240-cm^{-1} resonance.

In the model of the reflectance edge is caused by a zero crossing of ϵ_1. We hereafter refer to the frequency of the reflectance edge as the plasma frequency. A second, higher plasma frequency is, of course, to be expected at near-infrared or visible wavelengths. The temperature dependence of the reflectance may be qualitatively understood as follows. At $T=0$ the maximum number of carriers is condensed into the dissipationless superconducting state and the system can screen electromagnetic radiation effectively for frequencies as high as the low-temperature plasma frequency of 80 cm^{-1}. As the temperature is increased toward T_c, the fraction of carriers in the superconducting state decreases with the temperature dependence of the order parameter, decreasing the ability of the free carriers to screen and lowering the plasma frequency until it reaches zero for $T>T_c$. The broadening of the reflectance edge with increasing temperature arises from finite dissipation for $\omega<2\Delta(T)$ due to quasiparticles excited across the superconducting gap.

The plasmon hypothesis as implemented in our model can also account for the scaling of the frequency of the reflectance edge with T_{co}. We have observed a clear bump in the reflectance of $La_{1.85}Ca_{0.15}CuO_4$ near 250 cm^{-1}, indicating at least one strong phonon similar to that observed in the Sr-doped material. If we assume that the free-carrier density and relaxation times in $La_{1.85}Ca_{0.15}CuO_4$ and $La_{1.85}Sr_{0.15}CuO_4$ are comparable, and that the energy gap scales with the transition temperature, then the oscillator strength condensed into the superconducting state should also scale with transition temperature and so should the frequency of the reflectance edge.

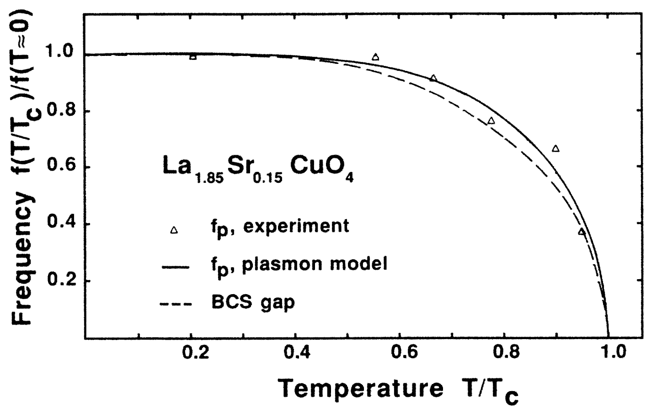

FIG. 4. Frequency of the reflectance edge vs reduced temperature for experiment (triangles) and for the plasmon model (solid line). The BCS gap is also plotted (dashed line). All quantities are normalized to 1 at $T=0$.

Although the plasma frequency in our model depends critically on the parameters of the normal-state fit, the scaling of the plasma frequency with temperature does not. Given the uncertainties inherent in modeling the normal-state reflectance of polycrystalline $La_{1.85}Sr_{0.15}CuO_4$, we do not attempt to make more than a rough comparison between the plasma frequencies in the model and in the experiment, and we are pleased that they agree to within 30%. Agreement could clearly be improved by adjusting the model parameters or the magnitude of the energy gap. However, our conclusions about the scaling of the plasma frequency with temperature and with T_c are robust. These conclusions depend only on the normal state being characterized by a low free-carrier density and a resonance that has large enough oscillator strength to yield a positive ϵ_1 for low FIR frequencies. Thus our main results should be valid for both the long-wavelength effective-medium characterization of the normal state which we have adopted, and for the short-wavelength approximation adopted by Schlesinger *et al.*[16]

LOW-FREQUENCY REFLECTANCE

Finally, we investigate the scaling of the reflectance with temperature at submillimeter wavelengths. We give the reflectance in a manner that is independent of the details of the fit to the normal state and independent of small (1–2%) temperature-independent losses due to surface scattering or to a normal surface layer. In Fig. 5 we have plotted $[R(T)-R_N]/(R_S-R_N)$ averaged over a 2-cm^{-1} band about 16 cm^{-1} for our experiment (squares) and our model of the plasmon hypothesis solid line. R_N and R_S are here the reflectance at $T=37$ and 24 K. The agreement between model and experiment is quite good. A model dielectric function which does not include the

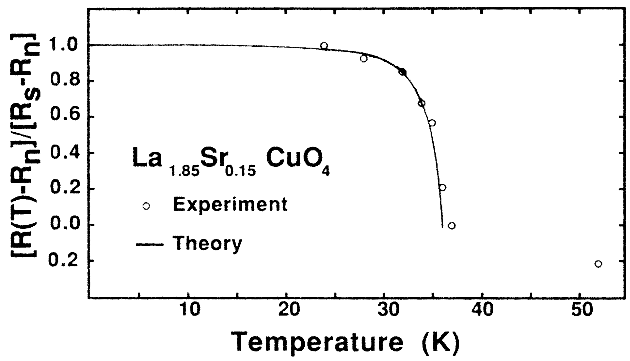

FIG. 5. Excess submillimeter reflectance $[R(T)-R_n]/(R_s-R_n)$ in the superconducting state vs temperature. Here, R_s and R_n were measured at 24 and 37 K, respectively. In this frequency range, the predictions of the plasmon model and those of the simple Mattis-Bardeen expressions are indistinguishable. Both models (solid line) agree with the experimental results (circles).

240-cm^{-1} phonon (consistent with the energy-gap hypothesis) gives results identical to those of the plasmon hypothesis. This indicates that the simple Mattis-Bardeen model of the superconducting state, which is equivalent to a two-fluid model with current carried by normal and superconducting carriers, is adequate to describe the temperature dependence of the reflectance at submillimeter wavelengths. This temperature dependence cannot be used to distinguish between the plasmon and the energy-gap hypotheses.

In order to assess the feasibility of using high-T_c superconductors for the construction of electronic devices that operate at submillimeter wavelengths, it is useful to compare the losses in these materials for frequencies less than $2\Delta_{BCS}/4 \approx 0.9 k_B T_c$ (where our calculations should be valid) to the losses in a good metal like copper. The absorption A_{Cu} of copper in the FIR was calculated using the dc conductivity and the Hagen-Rubens relation. The absorption for $T > T_c$ of the $La_{1.85}Sr_{0.15}CuO_4$ sample discussed here is typical for sintered polycrystalline samples,[11] and is roughly 200 times that of copper, independent of frequency to within 10% for frequencies less than $2\Delta_{BCS}/4 \approx 25$ cm^{-1}. Assuming no extrinsic surface losses and a temperature dependence of the absorption in the superconducting state described by BCS, $La_{1.85}Sr_{0.15}CuO_4$ would have to be cooled to $T/T_{co} \approx 0.4$ ($T \approx 14$ K) before its absorption equaled that of room-temperature copper for frequencies less than 25 cm^{-1}. A recent measurement[18] of the reflectance of an epitaxial film of $Y_1Ba_2Cu_3O_7$ ($T_c \approx 90$ K) showed a normal-state absorption of roughly $60 A_{Cu}$ for frequencies less than 200 cm^{-1}. If the temperature-dependent electrodynamics of this material for frequencies much less than the energy gap are also described by BCS, we estimate that this film would have to be cooled to $T/T_{co} \approx 0.5$ (45 K) before its absorption equalled that of room-temperature copper for frequencies less than $2\Delta_{BCS}/4 \approx 60$ cm^{-1}. Thus it appears that for existing materials, devices made from high-T_c superconducting oxides will have to be cooled to temperatures lower than $T_c/2$ in order to have lower losses than good metals. Some improvement in the conductivity of single crystals and epitaxial films can be expected.

CONCLUSION

The reflectance of the high-T_c superconductor $La_{1.85}Sr_{0.15}CuO_4$ for frequencies less than 100 cm^{-1} is well described by BCS electrodynamics. Our results are therefore consistent with a mean-field pairing theory of superconductivity. We have also shown that the temperature dependence of the FIR reflectivity cannot be used to distinguish between the plasmon hypothesis and the energy-gap hypothesis. The success of our model strengthens the plasmon hypothesis. However, a definitive understanding of the FIR dielectric function for $La_{1.85}Sr_{0.15}CuO_4$ must await a better understanding of the effect of crystal anisotropy on the reflectance of polycrystalline samples. Until this matter is clarified, it is premature to deduce the magnitude of the energy gap for this material from the infrared data.

ACKNOWLEDGMENTS

We wish to thank Professor A. Stacy for providing us with the samples used in this study. We also wish to thank Professor M. L. Cohen, Dr. G. A. Thomas, and P. E. Sulewski for helpful discussions. This research was supported by the Director, Office of Energy Research, Office of Basic Sciences, Materials Sciences Division of the U.S. Department of Energy, under Contract No. DE-AC03-76SF00098 (P.L.R.), and National Science Foundation Grant No. DMR-8400041 (A.Z.). One of us (A.Z.) acknowledges support from the Alfred P. Sloan Foundation, and another (M.S.S.) from AT&T Bell Laboratories.

[1]D. Esteve, J. M. Martinis, C. Urbina, M. H. Devoret, G. Collin, P. Monod, M. Ribault, and A. Revcolevschi, Europhys. Lett. 3, 1237 (1987); W. R. McGrath, H. K. Olsson, T. Claeson, S. Eriksson, and L.-G. Johannson, Europhys. Lett. 4, 357 (1987).

[2]L. C. Bourne, M. F. Crommie, A. Zettl, Hans-Conrad zur Loye, S. W. Weller, K. J. Leary, A. M. Stacy, K. J. Chang, M. L. Cohen, and D. E. Morris, Phys. Rev. Lett. 58, 2337 (1987); B. Batlogg, R. J. Cava, A. Jayaraman, R. B. van Dover, G. A. Kourouklis, S. Sunshine, D. W. Murphy, L. W. Rupp, H. S. Chen, A. White, K. T. Short, A. M. Mujsce, and E. A. Rietman, *ibid.* 58, 2333 (1987); L. C. Bourne, A. Zettl, T. W. Barbee III, and M. L. Cohen, Phys. Rev. B 36, 3990 (1987).

[3]T. A. Faltens, W. K. Ham, S. W. Keller, K. J. Leary, J. N. Michaels, A. M. Stacy, Hans-Conrad zur Loye, D. E. Morris, T. W. Barbee III, L. C. Bourne, M. L. Cohen, S. Hoen, and A. Zettl, Phys. Rev. Lett. 59, 915 (1987); B. Batlogg,

G. Kourouklis, W. Weber, R. J. Cava, A. Jayaraman, A. E. White, K. T. Short, L. W. Rupp, and E. A. Rietman, *ibid.* **59**, 912 (1987).

[4]V. J. Emery, Phys. Rev. Lett. **58**, 2794 (1987).

[5]J. R. Kirtley, C. C. Tsuei, S. I. Park, C. C. Chi, J. Rozen, and M. W. Shafer, Phys. Rev. B **35**, 7216 (1987); M. E. Hawley, K. E. Gray, D. W. Capone II, and D. G. Hinks, *ibid.* **35**, 7224 (1987); S. Pan, K. W. Ng, A. L. de Lozanne, J. M. Tarascon, and L. H. Greene, *ibid.* **35**, 7220 (1987).

[6]M. F. Crommie, L. C. Bourne, A. Zettl, M. L. Cohen, and A. Stacy, Phys. Rev. B **35**, 8853 (1987); J. Moreland, J. W. Ekin, L. F. Goodrich, T. E. Capobianco, A. F. Clark, J. Kwo, M. Hong, and S. H. Liou, *ibid.* **35**, 8856 (1987); J. R. Kirtley, R. T. Collins, Z. Schlesinger, W. J. Gallagher, R. L. Sandstrom, T. R. Dinger, and D. A. Chance, *ibid.* **35**, 8846 (1987).

[7]P. W. Anderson, in *Novel Superconductivity,* Proceedings of the International Workshop on Novel Mechanisms of Superconductivity, Berkeley, 1987, edited by S. A. Wolf and V. Z. Kresin (Plenum, New York, 1987), p. 295.

[8]U. Walter, M. S. Sherwin, A. Stacy, P. L. Richards, and A. Zettl, Phys. Rev. B **35**, 5327 (1987).

[9]P. E. Sulewski, A. J. Sievers, S. E. Russek, H. D. Hallen, D. K. Lathrop, and R. A. Buhrmann, Phys. Rev. B **35**, 5330 (1987); P. E. Sulewski, A. Sievers, R. Buhrman, J. Tarascon, and L. Greene, *ibid.* **35**, 8829 (1987).

[10]Z. Schlesinger, R. T. Collins, and M. W. Shafer, Phys. Rev. B **35**, 7232 (1987).

[11]G. A. Thomas, R. N. Bhatt, A. J. Millis, R. J. Cava, and E. A. Rietman, Proceedings of the Eighteenth International Conference on Low Temperature Physics, Kyoto, 1987 [Jpn. J. Appl. Phys. **26**, Suppl. 26-3, 1001 (1987)]. In this work the reflectance in the superconducting state was fit to a long-wavelength effective-medium theory. A value of the energy gap consistent with weak-coupling BCS theory was found to give the best fit.

[12]D. A. Bonn, J. E. Greedan, C. V. Stager, and T. Timusk, Solid State Commun. **62**, 383 (1987).

[13]D. A. Bonn, J. E. Greedan, C. V. Stager, T. Timusk, M. G. Doss, S. L. Herr, K. Kamaras, C. D. Porter, D. B. Tanner, J. M. Tarascon, W. R. McKinnon, and L. H. Greene, Phys. Rev. B **35**, 8843 (1987).

[14]G. A. Thomas, H. K. Ng, A. J. Millis, R. N. Bhatt, R. J. Cava, E. A. Rietman, D. W. Johnson, Jr., G. P. Epinosa, and J. M. Vändenberg, Phys. Rev. B **36**, 846 (1987).

[15]G. A. Thomas (private communication).

[16]Z. Schlesinger, R. T. Collins, M. W. Shafer, and E. M. Engler, Phys. Rev. B **36**, 5275 (1987).

[17]J. M. Bassat, P. Odier, and F. Gervais, Phys. Rev. B **35**, 7224 (1987).

[18]R. T. Collins, Z. Schlesinger, R. H. Koch, R. B. Laibowitz, T. S. Plaskett, P. Freitas, W. J. Gallagher, R. L. Sandstrom, and T. R. Dinger, Phys. Rev. Lett. **59**, 704 (1987).

[19]J. Orenstein, G. A. Thomas, D. H. Rapkine, C. G. Bethea, B. F. Levine, R. J. Cava, E. A. Rietman, and D. W. Johnston, Jr., Phys. Rev. B **36**, 729 (1987).

[20]D. C. Mattis and J. Bardeen, Phys. Rev. **111**, 412 (1958).

[21]See, for example, M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions,* National Bureau of Standards (U.S.) Applied Mathematics Series No. 55 (U.S. GPO, Washington, DC, 1970), formulas 25.4.37 and 25.4.39.