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Far-infrared measurement of $\alpha^2(\omega)F(\omega)$ in superconducting $La_{1.84}Sr_{0.16}CuO_{4-y}$

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A measurement of the far-infrared surface resistance for La-Sr-Cu oxide both above and below the superconducting transition temperature has been used to obtain the frequency dependence of $\alpha^2(\omega)F(\omega)$ in the superconducting state. The dominant feature in the spectrum is a single elastic or quasielastic peak near the gap energy.

Holstein[1] first showed that the coupling between electrons and phonons in metals allows an infrared absorption process at low temperatures associated with phonon generation by the excited electrons. The measurements by Joyce and Richards[2] on the far-infrared (FIR) absorptivity of superconducting and normal lead stimulated theoretical[3-5] and experimental[6-9] efforts to determine $\alpha^2(\omega)F(\omega)$, the product of the square of the electron-phonon matrix element and the phonon density of states in metals. The determination of $\alpha^2(\omega)F(\omega)$ by far-infrared means is now fairly well understood,[9] but the technique has had little impact because electron tunneling usually is a more accurate and simpler method for extracting the same information.[10] However, the highly textured surface of the sintered La-(Ba,Sr)-Cu oxide high-T_c superconducting compounds[11-16] makes tunneling measurements difficult and provides an opportunity to apply the alternate approach.

In this Rapid Communication we describe a far-infrared measurement of $\alpha^2(\omega)F(\omega)$ for superconducting $La_{1.84}Sr_{0.16}CuO_{4-y}$, a compound which has a sharp resistive transition at 38.5 K. The superconducting energy gap is measured to be 73.7 cm^{-1} so that $2\Delta/k_BT_c=2.7$, a value smaller than the Bardeen-Cooper-Schrieffer (BCS) value in agreement with earlier FIR measurements on related compounds.[17,18] Above this gap frequency extra FIR absorption is found in the superconducting state as required by the Holstein process. The frequency dependence of these data indicates that elastic or quasielastic scattering gives the dominant component in $\alpha^2(\omega)F(\omega)$, and the contribution from phonon excitations at larger frequencies is comparable with that found in Pb. Application of a general sum rule to the results demonstrates that other excitations outside of the measured frequency region do not contribute appreciably to $\alpha^2(\omega)F(\omega)$.

Two rectangular bar samples of $La_{1.84}Sr_{0.16}CuO_{4-y}$, $6\times3\times1$ mm^3, prepared at Bell Communications Research and described in Ref. 16 have been used in a two-bounce reflectivity measurement. These samples were chosen for this investigation because they show a particularly sharp resistive transition at 38.5 K with a width of 2 K. A summary of the FIR experimental technique has been given in Ref. 19.

The temperature dependences of the dc resistivity and also the square of the FIR reflectivity for a broad spectral distribution centered at 9 cm^{-1} with full width at half maximum (FWHM) of 9 cm^{-1} are shown in Fig. 1. The similar temperature dependences in the two sets of curves demonstrate that the dc and far-infrared probes are moni-

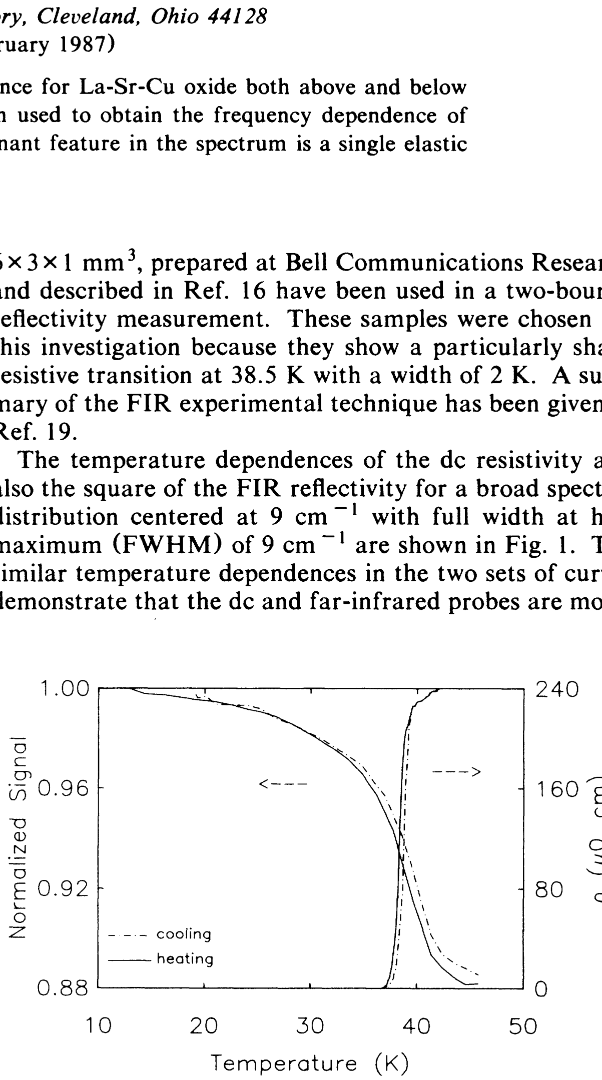

FIG. 1. The normalized integrated intensity after two reflections plotted as a function of temperature on heating and cooling the sample. The peak frequency is 9 cm^{-1} and FWHM 9 cm^{-1}. Comparing these curves with the dc resistivity data (right axis) for this sample on heating and cooling demonstrates that both probes see the same material.

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toring the same type of material.

The square of the frequency-dependent reflectivity at an angle of incidence of 45° is shown in Fig. 2(a) for both the superconducting (sample $T=4.2$ K) and normal state (sample $T=41$ K). The two curves at lower frequencies were taken using a lamellar grating interferometer while a Michelson interferometer was used for the two higher-frequency curves. The resolution of the curves is approximately 5% of the maximum frequency of each curve. Although the frequency dependence of the normal-state reflectivity is similar to that expected for a metal, the magnitude is much smaller over the measured frequency region than would be predicted from $\rho_{\mathrm{dc}}(41\ \mathrm{K})=200$ $\mu\Omega$ cm and the classical skin effect. This discrepancy presumably occurs because the samples contain a large number of voids which trap some of the incident radiation and enhance the absorptivity in the nonsuperconducting material. For the superconducting sample the reflectivity is larger than for the normal metal at frequencies less than 70 cm^{-1} and smaller above that value.

It requires some care to relate these reflectivity results to the effective surface impedance of the material. For typical metals, in which the reflectivities are near one, the normal incident reflectivity, absorptivity, and surface impedance are simply related:

$$R=1-A=1-4r\ , \qquad (1)$$

where r is the real part of the surface impedance normalized to the impedance of free space $(z=r+ix)$. However, because the absorptivity is so large in the high-frequency region in Fig. 2(a), we must use the complete Fresnel expressions to obtain the higher-order corrections to r. The Fresnel equations assume a knowledge of both the ratio of the imaginary to real part of the surface impedance $(|x|/r)$ and the reflectivity. As a good first approximation we have assumed the Hagen-Rubens relation that $|x|/r=1$, along with the measured reflectivity in the Fresnel equations to obtain the values of r_N and r_S shown in Fig. 2(b).

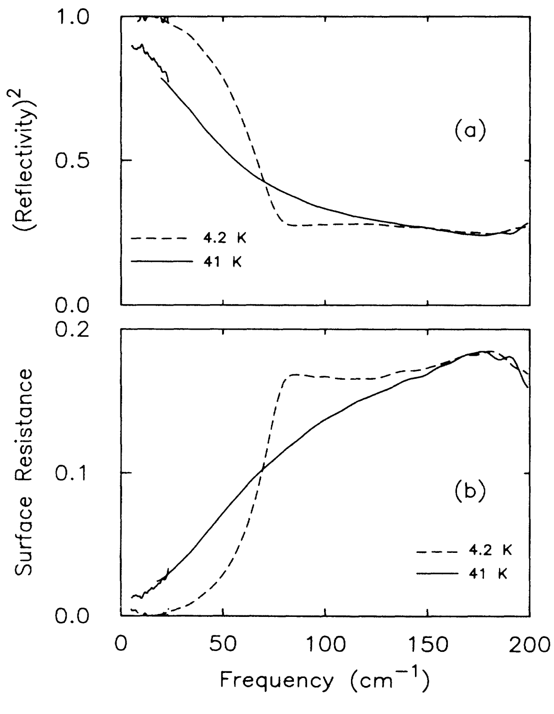

FIG. 2. (a) The reflectivity after two reflections at 45° angle of incidence with the samples at 4.2 and 41 K. (b) The surface resistance, r, obtained from the data in (a) as described in the text.

Two different ways of presenting the experimental results are shown in Fig. 3. Figure 3(a) gives the ratio of r_S/r_N, an accurate experimental quantity which clearly distinguishes the region where $r_S/r_N<1$. Because the material is a composite, the effective r_S is not zero below the superconducting energy gap. Figure 3(b) shows the difference between the two quantities (r_S-r_N). Those regions of the sample which do not contribute to the superconductivity do not appear in this plot. This plot also permits us to make use of the sum rule on (r_S-r_N) which states that[7]

$$\int (r_S-r_N)d\omega=0\ . \qquad (2)$$

Since the areas above and below the horizontal axis in Fig. 3(b) nearly cancel in the 0 to 200 cm^{-1} frequency region, the sum rule is exhausted, and hence there can be no ma-

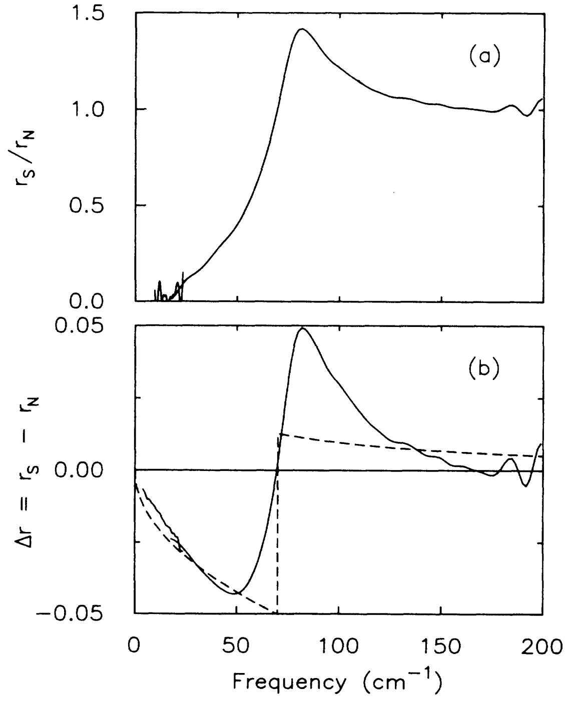

FIG. 3. (a) The ratio of the superconducting-state surface resistance to the normal-state surface resistance. (b) The difference of the superconducting and normal-state surface resistances. The contribution to the absorptivity from regions of the sample which do not go superconducting does not appear in this quantity. Also, structure due to surface morphology or inequivalence of sample mounting and reference mirror mounting is minimized. The dashed curve is a fit to a theory for elastic impurity scattering as described in the text.

jor contribution at higher frequencies to the $\alpha^2(\omega)F(\omega)$ determined in this restricted frequency interval.

To relate the frequency-dependent surface resistance to $\alpha^2(\omega)F(\omega)$ we follow Ref. 3. For elastic impurity scattering the scattering frequency in the normal state is $1/\tau_{N,i}$ and in the superconducting state for $\omega\tau > 1$ it is

$$1/\tau_{S,i} = 1/\tau_{N,i} E\{[1-(2\Delta/\omega)^2]^{1/2}\} \ , \tag{3}$$

where E is the complete elliptic integral of the second kind. The dashed curve in Fig. 3(b) shows a fit of Eq. (3) to the data (assuming $\omega\tau < 1$ and $\rho_{dc} = 4300\ \mu\Omega$ cm in the normal state). The gap energy is taken as the frequency, where $r_S = r_N$. The frequency dependence of the elastic scattering approximation is similar to the data but not correct in detail.

The scattering frequency produced by the electron-phonon coupling for a normal metal is given by[3]

$$1/\tau_N = (2\pi/\omega)\int d\Omega(\omega-\Omega)\alpha_{tr}^2 F(\Omega) \ , \tag{4}$$

where $\alpha_{tr}^2 F$ is the phonon density of states weighted by the amplitude for large-angle scattering on the Fermi surface. It is closely related to $\alpha_0^2 F$ which is measured in superconducting tunneling.[3] The relaxation frequency in the superconducting state has a related form but the frequency scale is shifted since an energy 2Δ must go to break up the Cooper pair. Differentiating the difference of the relaxation rates with respect to frequency gives[9]

$$\partial/\partial\omega(1/\tau_S - 1/\tau_N) = (\pi^2 2\Delta/\omega)\alpha_{tr}^2(\omega-2\Delta)F(\omega-2\Delta) + (\text{smoothly varying integral}) \ . \tag{5}$$

Farnworth and Tinusk[9] have shown that for pure Pb, structure in the derivative does correspond to structure in $\alpha_{tr}^2 F$, while Brandli[7] showed for a Pb-In alloy that both an elastic scattering and a phonon scattering part could be identified. One important difference between the FIR electrodynamics of low-temperature La-Sr-Cu oxide and Pb is that in the former case the sintered sample must satisfy $\omega\tau \ll 1$ over the entire frequency interval so that $(1/\tau) \propto r^2$, whereas in the latter case $\omega\tau \gg 1$ and $(1/\tau) \propto r$.

The experimental data are displayed as $r_S^2 - r_N^2$ in Fig. 4(a) [the dashed curve is the elastic scattering fit carried over from Fig. 3(b)] and the derivative of these data taken in Fig. 4(b). The determination of $r_S^2 - r_N^2$ requires that $r_S + r_N$ be used even though a component from nonsuperconducting material now may be present. Since r_N is smoothly varying the fact that it does not follow the Hagen-Rubens relation over the entire frequency interval will not be of major importance in the derivative spectrum.

The result in Fig. 4(b) shows that the dominant feature in $\alpha_{tr}^2 F$ is a single narrow peak centered at 73.7 cm^{-1}. This peak may be associated with a large elastic scattering component produced by the poor quality of the conductor, and the width could stem from a distribution of energy gaps caused by sample inhomogeneity. The center position would give an accurate value for the mean-gap energy, and hence $2\Delta/k_B T_c = 2.7$. If so, then the $\alpha_{tr}^2 F$ which we have measured would not be an intrinsic property of the superconducting material and better sample homogeneity should reduce the width and magnitude of this component.

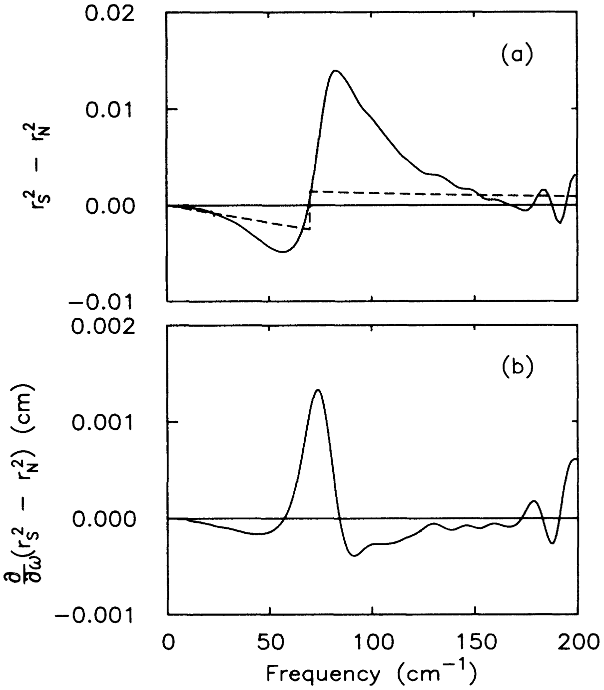

FIG. 4. (a) The difference of the square of the surface resistances as a function of frequency. For these samples $\omega\tau \ll 1$, so this quantity is proportional to the difference of the superconducting and normal-state relaxation rates. The dashed curve is obtained from the same parameters as the dashed curve in Fig. 3(b). (b) The derivative with respect to frequency of the curve in (a). This quantity is directly proportional to $\alpha_{tr}^2(\omega-2\Delta)F(\omega-2\Delta)$, apart from slowly varying contributions.

The other possibility is that the $\alpha_{tr}^2 F$ peak in Fig. 4(b) is associated with quasielastic scattering of the carriers by a low-frequency mode representing other degrees of freedom of the system. In this case, better quality material could show a sharper peak but its strength would not change. In either event the two small peaks in Fig. 4(b) centered at 100 and 130 cm^{-1} have magnitudes comparable to the phonon scattering contributions observed for Pb.[9]

In conclusion, the measurement of the real part of the surface impedance in the far infrared of a La-Sr-Cu oxide sample which shows a sharp resistive transition to the superconducting state has been used to identify $\alpha_{tr}^2 F$ for this high-temperature superconductor. Application of the sum rule to the surface resistance in the two states demonstrates that our measurement covers the important frequency region. Unlike pure metals or alloys where $\alpha_{tr}^2 F$ has a strong component which mimics the phonon density of states, here the dominant feature in the spectrum is a single elastic or quasielastic peak near the gap energy.

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