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# Far-infrared measurement of the gap of the high-$T_c$ superconductor $La_{1.85}Sr_{0.15}CuO_{4-x}$

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We report on far-infrared measurements of the energy gap of the high-temperature superconductor $La_{1.85}Sr_{0.15}CuO_{4-x}$. From the reflectance, measured over a wide range of frequencies, we calculate the optical conductivity. It shows phononlike lines at 239, 355, and 494 $cm^{-1}$ and a gaplike depression below 140 $cm^{-1}$. The strong edge seen in reflectance at 50 $cm^{-1}$, and misidentified by a number of investigators as the energy gap, does not appear in the conductivity. It is due to a zero crossing of $\epsilon_1(\omega)$ caused by the strong phonons.

There have been several recent reports[1-4] of attempts to measure the energy gap of the high-$T_c$ superconductor $La_{1.85}Sr_{0.15}CuO_{4-x}$ by far-infrared techniques. All were based on interpretations of reflectance spectra in analogy with the ordinary BCS metals where the reflectance in the superconducting state is unity up to the gap energy and then gradually falls to join the normal metal value well above the gap. In samples that displayed a large diamagnetic susceptibility in the superconducting state, unusually low values for the energy gap were obtained which implied values for the ratio $2\Delta/k_BT_c$, ranging from 1.8 to 2.4. The one reported measurement of the energy gap on a sample exhibiting a smaller diamagnetic susceptibility, also done by analysis of reflectance data, shows a gap to transition temperature ratio of $3.2 \pm 0.3$, close to the BCS value.[4] In contrast, much higher values come from tunneling measurements with the ratio ranging from 4 to 9.[5,6]

In this Rapid Communication we report on complete reflectance measurements from 30 to 950 $cm^{-1}$ above and below the transition temperature in $La_{1.85}Sr_{0.15}CuO_{4-x}$. With additional room-temperature measurements up to 21 000 $cm^{-1}$. we are able to perform accurate Kramers-Kronig analysis[7] and obtain the optical conductivity from 50 to 900 $cm^{-1}$. The data show that the infrared measurements are in fact consistent with a value of $2\Delta/k_BT_c$ that is greater than 3. The anomalously low values reported by Schlesinger, Collins, and Shafer,[1] by Sulewski, Sievers, Buhrman, and Tarascon[2] and by Walter *et al.*[3] result from a misinterpretation of the edge seen in reflectance spectra as the signature of the superconducting gap. This edge is not the microscopic gap but is instead related to a zero crossing of the real part of the dielectric response in a manner similar to the plasma edge in silver.[8] The actual gap is at higher frequency.

The preparation technique for the sample used here has been described previously. Samples prepared using this technique have a transition temperature which is stable over time and it has been suggested that any degradation of $T_c$ may be due to inhomogeneities.[9] Single reflection geometry near normal incidence was used. At the end of the experiment the sample was coated with a film; lead in the far-infrared experiments from 30 to 950 $cm^{-1}$ and aluminum for the near-infrared. Measurement of the reflectance of the metal-coated sample helps to eliminate effects of geometrical differences between the reference and the sample. This method allowed absolute determination of the reflectance of the sample to a precision of better than 0.5%.

Figure 1 shows the reflectance for $La_{1.85}Sr_{0.15}CuO_{4-x}$. Two temperatures are shown: one at 70 K above the transition temperature, the other well below it at 2 K. One of the most striking features is the plasmonlike edge at 50 $cm^{-1}$ at low temperatures that disappears almost completely at high temperatures. Our raw reflectance curve is in excellent agreement with previous data.[3]

Optical data are easier to interpret if the conductivity curve can be obtained from Kramers-Kronig transformations of the reflectance. In the normal state, the real part of the conductivity, shown in Fig. 2, has a constant background with a magnitude of about 100 $(\Omega\,cm)^{-1}$. From

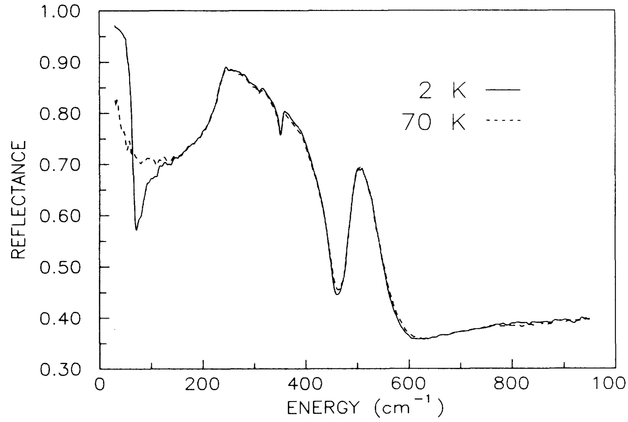

FIG. 1. Reflectance of $La_{1.85}Sr_{0.15}CuO_{4-x}$ in the normal state at 50–70 K (dashed curve) and in the superconducting state (solid curve).

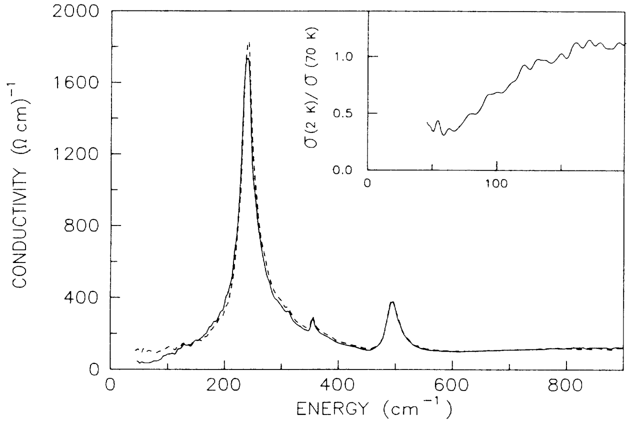

FIG. 2. The frequency-dependent conductivity of $La_{1.85}Sr_{0.15}CuO_{4-x}$ in the normal state (dashed curve) and in the superconducting state (solid curve). The inset shows the ratio of $\sigma(2\ K)/\sigma(70\ K)$ in the 0–200 cm$^{-1}$ region. We interpret the region of depressed conductivity below 80 cm$^{-1}$ as the BCS energy gap.

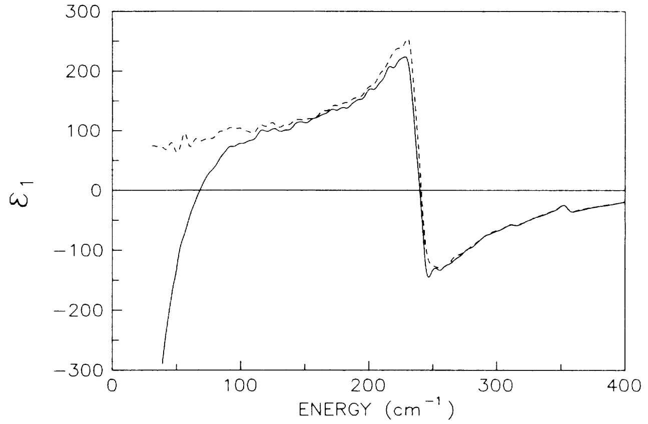

FIG. 3. The real part of the dielectric response of $La_{1.85}Sr_{0.15}CuO_{4-x}$, normal state (dashed curve), superconducting state (solid curve). We associate the shoulder seen in reflectivity at low frequency with the zero crossing of $\epsilon_1(\omega)$ at 60 cm$^{-1}$. It is caused by the presence of the strong phonon structure at 240 cm$^{-1}$.

the dc resistivity we would expect a conductivity of 2500 $(\Omega\,\mathrm{cm})^{-1}$, a discrepancy that may be due to the fact that this is a polycrystalline sample of a highly anisotropic material. dc measurements determine the resistance along the path of highest conductivity, whereas the optical data is an average over all crystal orientations. Superimposed on the constant conductivity are several prominent phonon peaks, a very strong one at 239, a weak peak at 355, and a medium sized one at 494 cm$^{-1}$. None of the peaks displays a particularly strong temperature dependence that would imply a strong coupling to the conduction electrons.

Below the transition temperature there is an overall depression of conductivity at low frequencies: The low-temperature conductivity is roughly 40% of the conductivity above the transition temperature, up to a frequency of 75 cm$^{-1}$ (10 meV). Above this frequency the two curves begin to approach one another and there is a crossing at 140 cm$^{-1}$, where the conductivity in the normal state exceeds the superconducting one. This depression of the low-frequency conductivity is seen clearly when one takes the ratio of the conductivity in the superconducting state to that in the normal state, as shown in the inset of Fig. 2. These observations are in qualitative agreement with a BCS electromagnetic response, as first described by Mattis and Bardeen.[10] However, the increase in conductivity above 75 cm$^{-1}$ is more gradual than is generally observed in superconductors and this may be due to the presence of a distribution of gaps that extends to higher frequencies from the 75-cm$^{-1}$ onset. An upper limit of 120 cm$^{-1}$ for the energy gap is reasonable as the superconducting conductivity meets the normal-state conductivity by 140 cm$^{-1}$. Thus, the far-infrared conductivity suggests an energy gap ranging from 75 to 120 cm$^{-1}$. This corresponds to a range for the ratio $2\Delta/k_BT_c$ of 2.9–4.5. The fact that the conductivity does not fall to zero below 75 cm$^{-1}$ indicates that either some of the sample is not superconducting, or that the energy gap is not apparent for some of the orientations of the grains in this polycrystalline sample.

In the real part of the conductivity there is no sign of the sharp structure seen in the reflectance at 50 cm$^{-1}$ in the superconducting state. The 50 cm$^{-1}$ shoulder arises from the zero crossing of the real part of the dielectric constant shown in Fig. 3. In a conventional superconductor there is no zero crossing of this quantity. $\epsilon_1(\omega)$ is usually large and negative and crosses zero at the plasma frequency in the ultraviolet. In the oxides, the phonon contribution to $\epsilon_1(\omega)$ raises the magnitude of $\epsilon_1(\omega)$ and causes it to cross zero at a very low frequency. The exact frequency depends on an interplay between several factors. It will move to lower frequencies if the magnitude of the conductivity singularity at the origin decreases. This happens as the order parameter of the transition is reduced by increased temperature as reported by Walter *et al.*[3]

In an ideal experimental geometry such as single crystal with polarized light, we expect the sample to become transparent in a narrow region between the zero crossing of $\epsilon_1(\omega)$ at 50 cm$^{-1}$ and the gap at 75 cm$^{-1}$. This region should move to lower frequencies as the order parameter is reduced by the imposition of a magnetic field or an increase of temperature. However, the verification of this conjecture must await the availability of suitable single-crystal samples.

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