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Energy gap in the high-T_c superconductor $La_{1.85}Sr_{0.15}CuO_4$

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The far-infrared (FIR) reflectance of the high-T_c superconductor $La_{1.85}Sr_{0.15}CuO_4$ has been measured in the frequency range 15 to 400 cm^{-1}. Below the superconducting onset temperature $T=36$ K, absorption features in the FIR spectrum indicate an energy gap with a functional form similar to the BCS gap, but with a smaller magnitude.

Very recently, intense scientific interest has focused on the layered perovskite-structure compounds La-*M*-Cu-O, with $M=$ Ba or Sr.[1-8] Of particular importance are the materials $La_{2-x}Ba_xCuO_4$ and $La_{2-x}Sr_xCuO_4$, with x typically 0.15. These systems show unusual transport and magnetic anomalies,[1-8] suggestive of superconducting transitions, with onset temperatures T_{c0} as high as 52 K under hydrostatic pressure[7] and 37 K under ambient pressure.[8]

Outstanding features of the La-Ba-Cu or La-Sr-Cu oxides include sharp drops in dc electrical resistivity below T_{c0}, with zero (or nearly zero) resistance states at lower temperature. In high-quality specimens, the transition width is as little as 1.4 K.[8] Similarly, magnetic susceptibility measurements indicate perfect diamagnetism throughout a substantial volume of the sample, as expected from the Meissner effect of a superconducting ground state.

The nature of the "superconducting" state in these compounds is of great current interest. Important questions address the behavior of the specific heat, the extent of "bulk" superconductivity and the capacity for persistent currents, strength of the electron-phonon coupling, and existence of an energy gap.

We have addressed the question of the energy gap in the anomalous superconducting phase of $La_{1.85}Sr_{0.15}CuO_4$ by means of far-infrared (FIR) reflectance measurements, over the frequency range 15–400 cm^{-1}. This spectral range is of interest since, in the BCS theory of superconductivity, a transition temperature of order 40 K would imply an energy gap of magnitude $2\Delta=(3.5-4)k_BT_c$, or the order of 100 cm^{-1}. In $La_{1.85}Sr_{0.15}CuO_4$, we find direct evidence that an energy gap exists. The gap first opens at $T_{c0}=36$ K, and displays a temperature dependence in rough agreement with the predictions of the BCS theory. The absolute magnitude of the gap (extrapolated to $T=0$) is, however, substantially smaller than the BCS prediction. We interpret our results in terms of a gapped superconducting ground state, and discuss possible mechanisms that might lead to a reduction of the gap magnitude.

We have prepared polycrystalline samples of $La_{1.85}Sr_{0.15}CuO_4$ by reaction of La_2O_3, CuO, and $SrCO_3$. Finely ground powders of the starting materials were pressed into a pellet approximately 1 cm in diameter, and sintered at 1100 °C for 44 h. The finished ceramiclike pellets were 78% dense. We have characterized our samples by careful magnetic susceptibility measurements, employing a SQUID magnetometer. During the measurement, the samples were cooled in a 16-G field. Figure 1 shows χ_g versus temperature for $La_{1.85}Sr_{0.15}CuO_4$. The sharp break in χ_g signals the onset of the superconducting transition at $T_{c0}=36$ K. The form of χ_g below T_{c0} suggests a transition

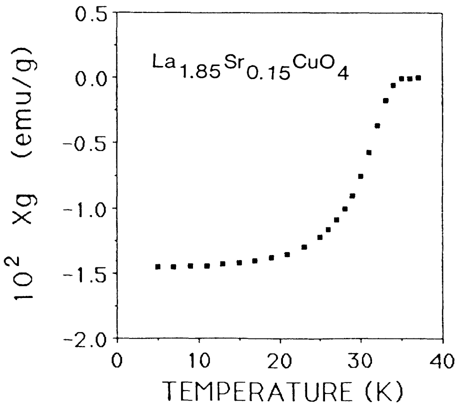

FIG. 1. Magnetic susceptibility of $La_{1.85}Sr_{0.15}CuO_4$ as a function of temperature. The initial break in the data determines $T_{c0}=36$ K.

width of approximately 10 K, indicating a reasonable sample quality. At low temperature, the *volume* susceptibility of our samples approaches $\chi_V = -8.0\times10^{-2}$ emu/cm^3. Within experimental error, this value is in exact agreement with that expected for a diamagnet: $\chi_V = -8.0\times10^{-2}$ emu/cm^3.

FIR reflectance measurements were performed with a Michelson interferometer, adapted to a helium-gas-flow cryostat to allow sample-temperature variations from room temperature to 6 K. During the experiment, chopped radiation impinged on the sample surface at near normal incidence. The reflected radiation was lock-in detected with a high-sensitivity 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.

Figure 2 shows a series of normalized reflectance spectra of $La_{1.85}Sr_{0.15}CuO_4$ at selected temperatures above and below $T_{c0}=36$ K. At 52 K, the reflectance R is approximately 86% at 20 cm^{-1}, and decreases smoothly with increasing frequency. Above 50 K, the reflectance curve was found to be rather insensitive to temperature. At 36 K, R has begun to rise at low frequency, and is approximately 90% at 20 cm^{-1}. At temperatures below approximately 34 K, the low-frequency reflectance is unity. In the low-temperature regime below T_{c0}, the reflectance follows a consistent behavior. At low frequency, R is near unity and decreases only slightly with increasing frequency. At moderate frequency, R displays a strong drop at a characteristic frequency f_0, and begins to flatten out once again at a higher characteristic frequency f_1. At 6 K, f_0 and f_1 are clearly identified at 50 and 66 cm^{-1}, respectively. We also note, at low temperatures, the presence of a shallow minimum in R near 70 cm^{-1}. Between 90 and 400 cm^{-1}, the reflectance decreases smoothly with increasing frequency, with no outstanding features.

Both f_0 and f_1 decrease with increasing temperature above 6 K. At and above 36 K, f_0 and f_1 are no longer identifiable. We associate the reflectance feature windowed by f_0 and f_1 in $La_{1.85}Sr_{0.15}CuO_4$ with the onset of photon absorption at the superconducting gap energy $h\nu = E_g = 2\Delta$. The absorption feature first appears just below $T_{c0}=36$ K. Below this critical temperature, the absorption edge at f_0 increases with decreasing temperature, as expected for an electronic gap that opens at T_{c0} and subsequently grows with decreasing temperature.

Figure 3 shows both f_0 and f_1 plotted versus temperature. Also plotted is f_p, which corresponds to the frequency of the (negative) peak in the frequency derivatives of the reflectance traces of Fig. 2 (the derivative traces are not shown). The derivative method yields a very accurate measurement of f_p, which lies between f_0 and f_1. The general behavior of the data of Fig. 3 is similar in form to that predicted by the BCS gap.[9] The solid line in Fig. 3 is the BCS gap, normalized to $T_c=36$ K, but with a zero temperature magnitude of $E_g=2\Delta=60$ cm^{-1}. This magnitude is approximately 30% *smaller* than that predicted from the expression[9]

$$2\Delta = 3.5k_BT_c\ , \qquad (1)$$

for which $E_g(T=0)=87$ cm^{-1} (using $T_c=T_{c0}=36$ K).

The relatively small magnitude of the zero-temperature gap we observe in $La_{1.85}Sr_{0.15}CuO_4$$(2\Delta/k_BT_{c0}=2.4)$ is surprising. The proportionality constant 3.5 in Eq. (1) is appropriate only to the weak-coupling limit, where $N(0)V \ll 1$, with $N(0)$ the density of states at the Fermi level and V the interaction potential. In the strong coupling limit, inelastic phonon processes give rise to quasiparticle damping which in turn *increases* the proportionality constant in Eq. (1).[10] Hence, in the strong coupling limit, Eq. (1) would tend to *underestimate* the gap magnitude, rather than overestimate it. In $La_{1.85}Sr_{0.15}CuO_4$, the Debye temperature of ≈ 400 (Ref. 11) and $T_c \approx 40$ K would suggest an electron-phonon coupling constant on the order of $g=0.43$. This is not too different from $g=0.39$ for lead, a strong coupling superconductor for which $E_g=4.1k_BT$.[12]

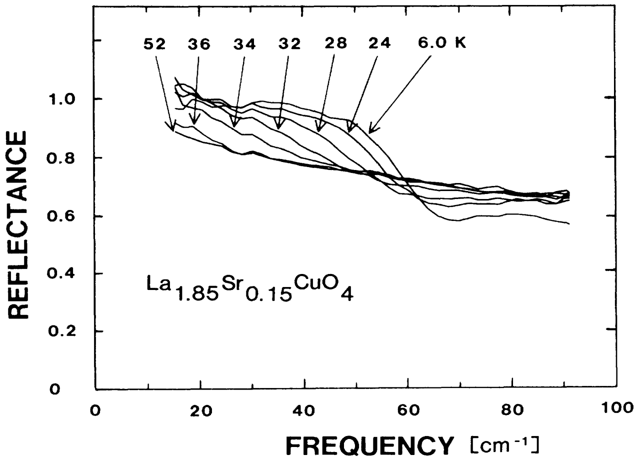

FIG. 2. Normalized reflectance of $La_{1.85}Sr_{0.15}CuO_4$ vs frequency at selected temperatures.

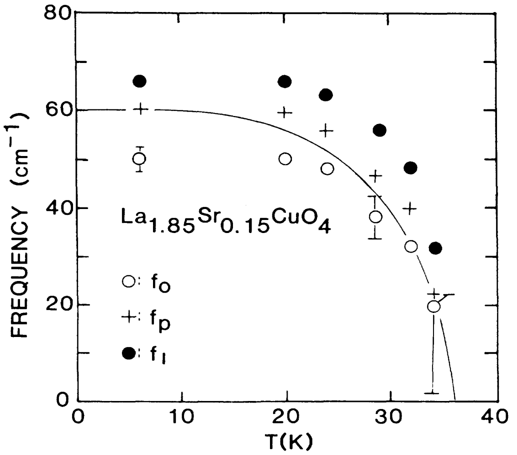

FIG. 3. Characteristic frequencies f_0, f_p, f_1 (see text) for FIR absorption in $La_{1.85}Sr_{0.15}CuO_4$, vs temperature. The solid line is the BCS gap, normalized to $T_c=36$ K, but reduced in magnitude by 30% from the prediction of Eq. (1).

We consider the possibility that the absorption we observe in $La_{1.85}Sr_{0.15}CuO_4$ results from a distribution of BCS-like gap energies within the sample. If a distribution were present, one would associate the upper energy cutoff with T_{c0} and the lower cutoff with T_{c1}, where T_{c1} represents the low-temperature limit of the transition width. In Fig. 1, we identify T_{c1} with the flattening out of χ at lower temperatures, hence $T_{c1} \approx 25$ K. A distribution would suggest that, at temperatures T such that $T_{c1} < T < T_{c0}$, a gap will exist at zero frequency. This is, however, in contrast to the FIR reflectance data of Fig. 2 which show, at $T = 24$ K, a well-developed absorption edge near 50 cm^{-1}, with no evidence for additional absorption at lower frequency. We do not, therefore, expect a gap distribution to be appropriate to $La_{1.85}Sr_{0.15}CuO_4$.

Another possibility for a small gap magnitude in a superconductor is a strongly variable density of states near the Fermi energy.[13] In $La_{1.85}Sr_{0.15}CuO_4$, one might expect the Fermi surface to result from hybridized copper and oxygen bands, with the possibility of density-of-states fluctuations. However, it remains uncertain if the mechanism of fluctuations in $N(E)$ alone would be sufficiently strong to reduce the gap to the observed value.

It is apparent that $La_{1.85}Sr_{0.15}CuO_4$ is not a conventional type-II superconductor. A number of novel mechanisms have been suggested[14,15] to account for the observed high-T_c anomalies in resistivity and susceptibility, including a superconducting ground state driven by a charge-density-wave instability,[14] and superconductivity with virtual plasmon exchange associated with the quasi-two-dimensional electronic structure.[15] Whether such mechanisms are consistent with the characteristic temperature dependence and magnitude of the superconducting gap we observe remains to be seen.

In conclusion, we have observed absorption features in the FIR spectrum of $La_{1.85}Sr_{0.15}CuO_4$ which identify an energy gap in the superconducting state. The development of the gap is consistent with the superconducting onset temperature $T_{c0} = 36$ K. On the other hand, the magnitude of the gap is inconsistent with the conventional strong (or weak) coupling theory of BCS superconductivity, and suggests novel interaction mechanisms in this unusual material. Complementary measurements of the magnetic field dependence of the gap, in addition to tunneling spectroscopy, should be of importance in understanding the details of the superconducting ground state. Such measurements are presently underway.

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