Charge Dynamics across the CuO_2 Planes in $La_{2-x}Sr_xCuO_4$

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Infrared reflectivity measurements on high-quality single crystals of $La_{2-x}Sr_xCuO_4$ were performed with polarization perpendicular to the CuO_2 planes (**E**$\parallel$**c**). The c-axis spectrum shows a dramatic change at T_c, from a featureless spectrum above T_c to the one with a sharp reflectivity edge below T_c. We find that the spectrum below T_c is dominated by the plasma edge arising from the carriers condensed in the superconducting state, not by a superconducting gap excitation, and that the superconducting transition is accompanied by the onset of coherent charge transport along the c axis.

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In contrast to the tremendous number of experimental studies made on the in-plane properties of the high-T_c superconductors, much less is known about the properties in the direction across the CuO_2 planes. This is partly because high-T_c crystals are not thick enough to obtain reliable experimental data with quantitative rigor. Reflecting this situation, many theoretical models are purely two dimensional, paying less attention to the c-axis properties. However, it is not so obvious whether the actual electronic state is two dimensional or not, and this has to be verified by investigating the c-axis properties in the normal state.

Concerning the superconducting state there remain unsettled subjects of debate even regarding the in-plane electromagnetic response of the high-T_c superconducting state, e.g., whether a BCS gap determines the optical response at $T < T_c$ [1], and whether or not there exists a coherence peak in the T dependence of the optical conductivity [2]. In this paper we report c-axis reflectivity spectra (with polarization perpendicular to the CuO_2 planes) for large high-quality single crystals of $La_{2-x}Sr_xCuO_4$ at three compositions, $x=0.10$ ($T_c=27$ K), $x=0.13$ ($T_c=32$ K), and $x=0.16$ ($T_c=34$ K). $La_{2-x}Sr_xCuO_4$ has one CuO_2 plane per unit cell and the results are free from complications arising from complex structure. While the spectrum above T_c is unusually featureless with a much depressed conductivity over a wide energy region, a sharp plasma edge of superconducting carriers appears at a frequency in the 20-50-cm^{-1} range in the spectrum below T_c. The result implies that the screened plasma frequency of superconducting carriers is smaller than the superconducting gap and as a consequence the optical response of the superconducting state is determined not by a gap but by the density of condensed carriers. This is quite an extraordinary situation that has never been met in other superconducting materials. The result also indicates that the superconducting transition is accompanied with the onset of coherent charge transport across the CuO_2 planes, which is blocked above T_c.

Crystals used in this study were grown by the traveling-solvent-floating-zone method [3]. The Sr composition is varied by following the phase diagram reported previously [4]. A single-domain crystal was grown in nearly cylindrical shape, 5 mm diameter and 50 mm length, with facets corresponding to the a-b surface. The cross section of the cylinder then contains the a and c axes and has sufficient area for the polarized optical measurements.

Homogeneity checks guaranteed the Sr composition within ± 0.01 over an entire crystal. The superconducting transition is sharp ($\Delta T_c < 2$ K) in both resistivity and SQUID magnetization measurements and the T_c value at each x coincides precisely with that reported for well-characterized ceramic samples [5].

Polarized optical measurements were done using a rapid-scan Fourier-type spectrometer in the infrared region (20–10000 cm^{-1}) and at temperatures between 8 and 300 K. The reflectivity was determined with accuracy 1% or less with a gold evaporated mirror as a reference.

The reflectivity spectra for **E**$\parallel$**c** are nearly flat above 1000 cm^{-1} for all the compositions, in contrast to the **E**$\perp$**c** spectra which are characterized by an edge at $\sim$6000 cm^{-1} and high reflectivity in the low-energy region. The c-axis spectra below 1000 cm^{-1} are dominated by two or more optical phonon bands centered at $\sim$300 cm^{-1} and at $\sim$600 cm^{-1}. However, a small electronic contribution is indicated by a slight rise of reflectivity below 100 cm^{-1}. The overall spectrum does not change appreciably when the sample is cooled down to a temperature just above T_c. Figures 1(a)–1(c) show the spectra below 360 cm^{-1} for $x=0.10$, 0.13, and 0.16, respectively.

A dramatic change in the spectrum is seen when the sample is in the superconducting state. A sharp reflectivity edge appears at frequency far below the lowest phonon band, and it becomes sharper as T is lowered. The edge frequency rapidly increases with x, and obviously does not scale with T_c. Note that the edge energy (ω_p) in units of k_BT_c is much lower than the BCS weak-coupling limit 3.5, e.g., $\hbar\omega_p/k_BT_c \simeq 0.9$ for $x=0.10$ and 1.7 for $x=0.16$. The spectral change is not so obvious in the a-b plane (**E**$\perp$**c**) spectrum which exhibits a gradual increase in R with decreasing T but no distinct feature of the superconducting transition—a broad weak feature is seen at 100 cm^{-1} ($\sim 5k_BT_c$) for $x=0.10$ only when we

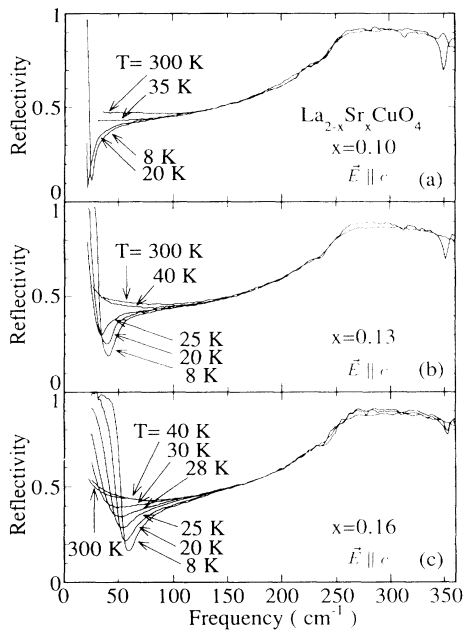

FIG. 1. Infrared reflectivity spectra of $La_{2-x}Sr_xCuO_4$ with polarization perpendicular to the CuO_2 planes (**E**∥**c**) above and below T_c for (a) $x=0.10$ ($T_c=27$ K), (b) $x=0.13$ ($T_c=32$ K), and (c) $x=0.16$ ($T_c=34$ K).

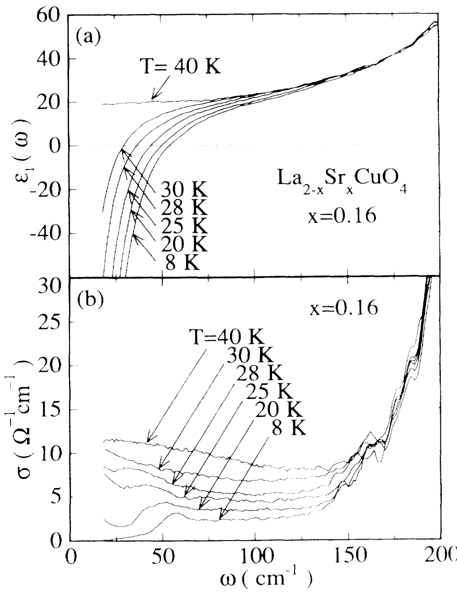

FIG. 2. (a) Real part of dielectric function and (b) optical conductivity of $La_{1.84}Sr_{0.16}Cu_4$ at various temperatures obtained from the Kramers-Kronig analysis on the c-axis reflectivity data. The optical phonon contribution centered at 240 cm^{-1} is truncated to emphasize the electronic contribution to the optical conductivity spectra.

take the normal-to-superconducting ratio of the reflectivity. It turns out from the present results that the earlier optical spectra measured on ceramic samples were dominated by the c-axis contribution [6–8].

The reflectivity data were transformed into the complex dielectric function $[\epsilon(\omega)=\epsilon_1(\omega)+i\epsilon_2(\omega)]$ by Kramers-Kronig (KK) analysis. Since we are concerned with the low-energy region below 200 cm^{-1}, the extrapolation of the reflectivity spectrum to the higher-energy region does not affect the result. However, the result somehow depends on the extrapolation to lower energies below 20 cm^{-1}. In the normal state we can apply the Hagen-Rubens formula for the low-energy extrapolation in order that the zero-frequency value of the conductivity $\sigma(\omega) =(1/4\pi)\omega\epsilon_2(\omega)$ coincides with the measured dc values. In the superconducting state it is critically important to decide how the reflectivity approaches unity. We adopted an extrapolation scheme suggested by van der Marel *et al.* [9] who assumed the low-frequency dielectric function to be

$$\epsilon(\omega)=\epsilon_\infty-\frac{\omega_{ps}^2}{\omega(\omega+i0^+)}-\frac{\omega_{p0}^2-\omega_{ps}^2}{\omega(\omega+i\gamma)}\,, \qquad (1)$$

where ϵ_∞ is the high-frequency dielectric constant, the second term is the contribution of the condensed carriers, giving rise to a δ-function peak at $\omega=0$ in $\sigma(\omega)$, and the last term represents the normal component formed by thermally excited quasiparticles having scattering rate γ $(=1/\tau)$ and density proportional to $\omega_{p0}^2-\omega_{ps}^2$. Equation (1) assumes that $\omega<2\Delta$, 2Δ being the superconducting gap, and the carrier density, represented by ω_{p0}, is the same below and above T_c. The internal consistency of these assumptions is discussed below.

The result of the KK transformation is shown in Fig. 2 for $x=0.16$ ($T_c=34$ K) which was investigated most extensively. The results for other compositions are essentially the same, so the analysis and discussion below are focused on $x=0.16$. In the normal state the conductivity is very low up to ~150 cm^{-1} but remains finite even when the temperature is lowered. However, the magnitude of the conductivity is only weakly ω dependent and orders of magnitude smaller than the conductivity in the a-b plane in the same frequency region. The conductivity is even smaller than the Mott minimum metallic conductivity ($\sigma_{\min}^c\sim100$ $\Omega^{-1}cm^{-1}$ [10]), thus suggestive of an unconventional charge transport in the sense that coherent transport is blocked across the CuO_2 planes as suggested by Anderson [11].

Actually, the T dependence of the dc resistivity is substantially different from the T-linear resistivity in the a-b plane. The anisotropic ratio ρ_c/ρ_{ab}, which is about 200 and T independent above 200 K, remarkably increases as T is lowered. The fact that $\sigma(\omega)$ is depressed at a very low level up to high frequencies rules out the possibility that the carriers are localized or trapped due to disorder or phonons in the LaO layers, and provides evidence for a two-dimensional electronic state for $T>T_c$. The low-frequency conductivity is nearly T independent in conjunction with a slowly decreasing dc resistivity.

Below T_c the conductivity is rapidly depressed and is dominated by thermally excited quasiparticles near T_c. At the lowest temperature (~ 8 K), $\sigma(\omega)$ is suppressed to a smaller magnitude which indicates that most of the spectral weight in the normal state collapses into the δ function at $\omega = 0$. It is not clear whether the small conductivity surviving at 8 K is intrinsic or extrinsic in origin. Note that no definite gap is identified at frequencies corresponding to $\hbar\omega/k_BT_c \geq 3.5$, for $\omega \geq 90$ cm^{-1} for $T_c = 34$ K, at which the c-axis superconducting gap (2Δ) was identified in the Raman and tunneling spectra of Y-Ba-Cu-O (YBCO) or Bi 2:2:1:2 [12,13]. These observations are consistent with superconductivity in the clean limit ($\gamma < 2\Delta$) and moreover postulate a quite anomalous situation, $\omega_p = \omega_{ps}/\epsilon_\infty^{1/2} < 2\Delta$.

The Mattis-Bardeen formula usually applied to superconductors in the dirty limit requires both $\sigma = 0$ and $\epsilon_1 < 0$ for $\hbar\omega < 2\Delta$. A gap is then defined as the deviation from 100% reflectivity. This is not the case with the present spectra in the superconducting state. The frequency ω_0 at which a deviation from 100% reflectivity becomes apparent is $\hbar\omega_0/k_BT_c < 1.7$ for $x = 0.16$ and even smaller than 1 for other compositions, and does not scale with T_c. As shown in Fig. 2(a), ϵ_1 is positive and nearly constant (~ 25) below 100 cm^{-1} in the normal state and then changes sign below T_c, decreasing rapidly and following a $-1/\omega^2$ decay, characteristic of the superconducting state. We thus find the real part of the dielectric constant in the superconducting state is well approximated by $\epsilon_1^s(\omega) = \epsilon_\infty - \omega_{ps}^2/\omega^2$, consistent with the assumption of Eq. (1). The reflectivity edge is then determined by a zero crossing of $\epsilon_1(\omega)$, at $\omega_p = \omega_{ps}/\epsilon_\infty^{1/2}$, and corresponds to a screened plasma edge associated with the superconducting carriers. Therefore, the energy of the reflectivity edge does not define a gap but is determined by the density of superconducting carriers in the present case.

The dielectric function $\epsilon_1^s(\omega)$ is the one postulated by the London model in the clean limit. Relevant parameters describing the c-axis optical response at finite temperatures ($0 < T < T_c$) are thus $\omega_{ps}(T)$ and $\gamma(T)$. Both parameters are roughly estimated by a fit of the reflectivity spectrum using Eq. (1). The best-fit parameters are $\epsilon_\infty = 25$, $\omega_{p0} = 300$ cm^{-1}, $\omega_{ps} = 280$ cm^{-1}, and $\gamma = 2$ cm^{-1} at 8 K. At 30 K, just below T_c, the values of ω_{ps} and γ change to 120 and 75 cm^{-1}, respectively. The oscillator strength ω_{ps} corresponds to the "missing" area of the $\sigma(\omega)$ spectrum, i.e., $\int_{0^+}^{\infty}(\sigma_n - \sigma_s)d\omega$, as well as to the frequency of the zero crossing $\epsilon_1(\omega)$ ($\omega_{ps} = \omega_p\epsilon_\infty^{1/2}$). We actually obtain nearly equal values to within 20%. It is also found that ω_{ps} at 8 K is nearly identical to the normal-state oscillator strength ω_{pn} since $\int_{0^+}^{\infty}\sigma_s(\omega)d\omega$ is small at $T \ll T_c$, which gives further evidence for the superconductivity in the clean limit. The T dependence of ω_{ps} is shown in Fig. 3(a) on a normalized scale. The dotted and dashed curves represent the BCS and two-fluid models, respectively. Essentially the same c-axis response was reported by Collins *et al.* [14] and by Koch *et al.* [15] on 90-K YBCO. In this material a reflectivity edge in the superconducting state is observed at about 100 cm^{-1}. We further note that, considering that high-T_c superconductivity is presumably in the clean limit also in the a-b plane, the ir response of the a-b plane is similar to the c-axis one where the relevant parameter is the superfluid density rather than the energy gap. Interpretations along this line have been made on YBCO-related materials [16,17] and Bi 2:2:1:2 [18].

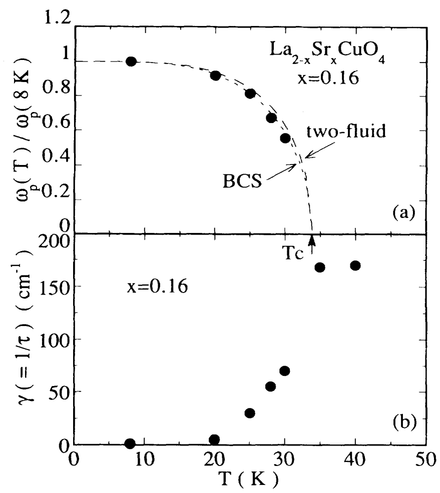

FIG. 3. (a) Plasma frequency of the superconducting carriers plotted as a function of temperature. Plasma frequencies are normalized to the lowest-temperature (8 K) value. The dotted and dashed curves represent the weak coupling BCS and the phenomenological two-fluid model, respectively. (b) Temperature dependence of the scattering rate of the carriers (quasiparticles) along the c axis.

The quasiparticle scattering rate $\gamma(T)$ is plotted in Fig. 3(b). The T dependence looks very similar to that for the quasiparticle scattering rate in the a-b plane as observed in Bi 2:2:1:2 [18] and YBCO [19], which is ascribed to the suppression of the scattering process responsible for the T-linear resistivity. However, the implication of the present result might be different. The value of γ in the normal state is of less significance—γ is estimated to be about 170 cm^{-1} through $\sigma(0) = \omega_{pn}^2/\gamma$ using the dc resistivity. The electronic conductivity is spread from $\omega = 0$ to about 300 cm^{-1} with values considerably reduced below the minimum metallic conductivity. In order to form a coherent metallic conductivity at $\omega = 0$ [$\sigma(0) \geq \sigma_{\min}^c$] by redistributing the total spectral weight $\omega_{pn} \sim 300$ cm^{-1}, $\sigma(\omega)$ has to have a narrow (Drude) peak at $\omega = 0$, with γ less than 15 cm^{-1}. In this case $\sigma(\omega)$ at the presumable superconducting gap $\omega = 2\Delta$ ($\sim 3.5k_BT_c \sim 90$ cm^{-1}) would be vanishingly small and it would be difficult to observe the energy gap in an infrared measurement since all of the area in $\sigma(\omega)$ collapses into the δ function at $\omega = 0$. This is basically what we have observed here. Therefore, we may conclude that the superconducting transition restores a coherent transport across the CuO_2 planes which is blocked above T_c. The rapid decrease of γ below T_c shown in Fig. 3(b) is a demonstration of the restoration of the coherent transport.

Although a quantitative analysis of the *a-b* spectra of $La_{2-x}Sr_xCuO_4$ is not free from large ambiguity, the muon-spin-relaxation (μSR) experiment provides us with the superfluid mass-density ratio related to the *a-b* plane. The μSR-estimated magnetic penetration for $x \sim 0.16$ is $\lambda_{ab} \sim 0.3$ μm well below T_c which corresponds to the plasma frequency $\omega_{pab} = 1/2\pi\lambda_{ab} \sim 5000$ cm^{-1} [20]. Then, the anisotropy $\omega_{pab}/\omega_{pc} \sim 17$ results. The square of this ratio, ~ 280, should be compared with the ratio of the normal-state resistivity ρ_c/ρ_{ab} which is much larger (≥ 500) below 200 K but is comparable (~ 200) at higher temperatures.

Finally, we emphasize two important points raised in the present study. First, the dramatic change of the spectrum below and above T_c is triggered by the restoration of the coherent charge transport along the *c* axis. In this regard, the superconducting transition is a dimensional crossover from two above T_c to three below T_c. The second is that the characteristic *c*-axis spectrum below T_c arises from the unprecedented situation $\omega_p = \omega_{ps}/\epsilon_\infty^{1/2} < 2\Delta$ which seems to be common with all the high-T_c cuprates. This condition is suggestive of a small interlayer hopping $t < \Delta$ in which case the system may be regarded as a "microscopic" Josephson array.

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[1] For a review, see D. B. Tanner and T. Timusk, in *Physical Properties of High Temperature Superconductors III*, edited by D. M. Ginsberg (World Scientific, Singapore, 1992), p. 363, and references therein.

[2] K. Holczer, L. Farro, L. Mihaly, and G. Grüner, Phys. Rev. Lett. **67**, 152 (1991); M. C. Nuss, P. M. Mankiewich, M. L. O'Malley, E. H. Westerwick, and P. B. Littlewood, *ibid.* **66**, 3305 (1991).

[3] I. Tanaka and H. Kojima, Nature (London) **337**, 21 (1989).

[4] S. Uchida *et al.*, Phys. Rev. B **43**, 7942 (1991).

[5] H. Takagi, T. Ido, S. Ishibashi, M. Uota, and S. Uchida, Phys. Rev. B **40**, 2254 (1989).

[6] L. Degiorgi, E. Kaldis, and P. Wachter, Solid State Commun. **64**, 873 (1987).

[7] M. S. Sherwin, P. L. Richards, and A. Zettl, Phys. Rev. B **37**, 1587 (1988).

[8] T. W. Noh, S. G. Kaplan, and A. J. Sievers, Phys. Rev. Lett. **62**, 599 (1989), these authors measured the sphere resonance for various Sr compositions. The present result confirms that the resonance takes place near the frequency corresponding to the reflectivity edge.

[9] D. van der Marel *et al.*, Physica (Amsterdam) **176C**, 1 (1991).

[10] T. Ito, H. Takagi, T. Ido, S. Ishibashi, and S. Uchida, Nature (London) **350**, 596 (1991).

[11] P. W. Anderson, Physica (Amsterdam) **185-189C**, 11 (1991); Science **256**, 1526 (1992).

[12] K. F. McCarty, J. Z. Liu, R. N. Shelton, and H. B. Radousky, Phys. Rev. B **42**, 9973 (1990).

[13] J. S. Tsai *et al.*, Physica (Amsterdam) **153-155C**, 1385 (1988).

[14] R. T. Collins, Z. Schlesinger, F. Holtzberg, and C. Feild, Phys. Rev. Lett. **63**, 422 (1989).

[15] B. Koch *et al.*, in *Electronic Properties of High-T_c Superconductors and Related Compounds,* edited by H. Kuzmay, M. Mehring, and J. Fink (Springer, Berlin, Heidelberg, 1990), p. 290.

[16] F. Gao *et al.*, Phys. Rev. B **43**, 10383 (1991).

[17] B. Bucher, J. Karpinski, E. Kaldis, and P. Wachter, Phys. Rev. B **45**, 3026 (1992).

[18] D. B. Romero *et al.*, Phys. Rev. Lett. **68**, 1590 (1992).

[19] D. A. Bonn, P. Dosanjh, R. Liang, and W. N. Hardy, Phys. Rev. Lett. **68**, 2390 (1992).

[20] Y. J. Uemura *et al.*, Phys. Rev. Lett. **62**, 2317 (1989).