Interplane Charge Dynamics in $La_{2-x}Sr_xCuO_4$

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We present c-axis polarized optical spectrum $\sigma_c(\omega)$ and out-of-plane Hall coefficient (R_H^c) in the normal state of $La_{2-x}Sr_xCuO_4$ as a function of doping. While $\sigma_c(\omega)$ is dominated by a Drude-like peak at $\omega = 0$ and R_H^c has finite negative values in the overdoped region, the low-energy $\sigma_c(\omega)$ is severely suppressed and nearly ω independent, and R_H^c is very small for underdoped compounds. The results provide evidence for different charge dynamics normal to the planes and clearly demonstrate how the charge dynamics changes with doping.

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The in-plane optical spectrum of the parent insulator of the high-T_c cuprates, such as La_2CuO_4, is characterized by the O2p-Cu3d charge-transfer (CT) excitation with a threshold energy in the 1.5–2.0 eV range. Doping, e.g., Sr substitution for La_2CuO_4, induces a dramatic change in the spectrum [1]. The spectral intensity of the CT excitation is rapidly transferred to low-energy excitations below $\sim$ 1.0 eV, which implies that the O2p and Cu3d states, initially separated by a CT gap, are redistributed upon doping and form new bands of states in the CT gap region. Similar transfer of the spectral weight has also been observed by electron-energy-loss (EELS) [2] and x-ray absorption (XAS) spectroscopy [3]. Such a spectral change, or rearrangement of the states, is a key signature of strongly correlated electrons [4–7]. Charge carriers (quasiparticles) are highly itinerant in the CuO_2 plane as evidenced from low in-plane resistivity (ρ_{ab}) [8,9] and a pronounced Drude peak in the optical conductivity spectrum [1].

An understanding of the electronic states in this anisotropic medium is not complete without an understanding of the out-of-plane charge dynamics. Recently, it has become possible to measure c-axis (perpendicular to the CuO_2 planes) properties reliably and accurately owing to the success in growing large high quality single crystals of the $La_{2-x}Sr_xCuO_4$ system over a wide compositional range [9]. It has been shown that the out-of-plane resistivity ρ_c has a negative T coefficient at low temperatures for $x \lesssim 0.15$ where the T coefficient of ρ_{ab} is positive. This fact suggests that the conduction mechanism might be different between the two directions. The most important finding about the out-of-plane transport is that ρ_c exhibits a remarkable decrease with dopant concentration [9]. This is a general trend of all the known high-T_c cuprates [10]. Therefore, the decrease of ρ_c with x signals a marked change in charge transport as the doping level increases.

In this paper we show the results of the c-axis polarized optical spectrum measured in the normal state of the $La_{2-x}Sr_xCuO_4$ single crystals in the compositional range $0 \le x \le 0.30$ which covers the insulating, superconducting, and overdoped metallic phases. We find an appreciable c-axis infrared conductivity of Drude form for an overdoped compound ($x = 0.30$), consistent with the metallic conduction at $\omega = 0$. Upon reducing the Sr composition and entering the superconducting regime, the low-energy spectral weight below $\sim$ 0.3 eV is suppressed very rapidly. The results demonstrate that the interplane conduction process is entirely different from the intraplane one and might be incoherent hopping between layers. We also examine the Hall effect by applying magnetic field parallel to the plane. The Hall coefficient (R_H^c) has finite negative values for the overdoped compound but it becomes very small for superconducting compositions.

$La_{2-x}Sr_xCuO_4$ single crystals were grown using the traveling-solvent-floating zone (TSFZ) method. Details in growth condition and characterization were described in Ref. [9]. The size of the grown crystal is typically 5 mmϕ×40 mm, so that measurements of the c-axis properties are easy and have an accuracy as high as those of the in-plane properties. In particular, we could prepare a c-axis long sample for the measurement of the Hall coefficient R_H^c with current flowing along the c axis. The c-axis polarized reflectivity spectra at room temperature were measured in the energy range between 0.005 and 40 eV. The infrared region was covered by a rapid-scan Fourier-type interferometer and the ultraviolet region by utilizing synchrotron radiation in the facility of the Institute for Solid State Physics, University of Tokyo. The high-energy spectrum [11] does not change with Sr composition, and hence is not of central concern in the present paper.

The c-axis optical conductivity $\sigma_c(\omega)$ is shown in Fig. 1 below 2 eV. $\sigma_c(\omega)$ is calculated from the reflectivity spectrum $R_c(\omega)$ using the Kramers-Kronig transformation in which the low-energy reflectivity is extrapolated to unity at $\omega = 0$ using the Hagen-Rubens relation so as to get $\sigma_c(0)$ in coincidence with the measured dc value [9]. The electronic contribution to $\sigma_c(\omega)$ of the undoped ($x = 0$) compound is absolutely vanishing up to $\sim$ 1 eV above which the interband transitions start. Upon doping some spectral weight is transferred into the low-energy region as in the case of the in-plane spectrum [1], but the trans-

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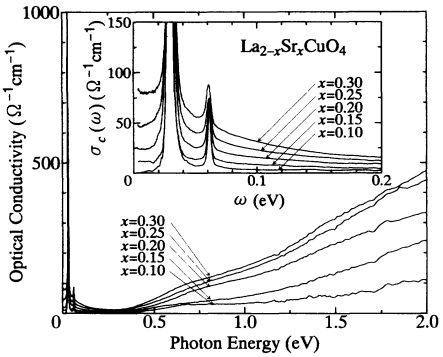

FIG. 1. Optical conductivity spectra below 2.0 eV of c axis $La_{2-x}Sr_xCuO_4$ for several x. Expanded spectra in the low-energy region are shown in the inset.

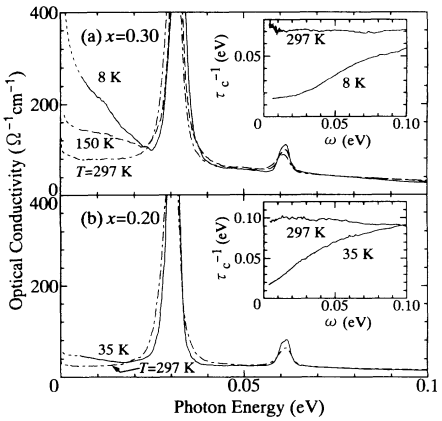

FIG. 2. c-axis optical conductivity spectra of the overdoped compound with $x = 0.30$ (a) and $x = 0.20$ (b) at different temperatures below 300 K. The inset of each figure shows ω dependence of the carrier scattering rate (τ_c^{-1}) obtained from the fit to the generalized Drude formula.

ferred weight is quite small, particularly below ~ 0.25 eV. The c-axis spectrum of doped material is similar to the in-plane spectrum in the lightly doped region in that it is decomposed into two parts, the Drude and midinfrared band [1]. An obvious difference is that development of a Drude conductivity is suppressed in the c-axis spectrum until the compound becomes heavily doped. The c-axis optical conductivity below 0.25 eV is nearly ω independent for the underdoped compounds ($x \leq 0.15$).

The expanded lower-energy part is shown in the inset of Fig. 1. The electronic contribution is superposed on three optical phonons at ~ 0.03, ~ 0.04, and ~ 0.06 eV. The two modes have considerable oscillator strengths; in particular the strength of the 0.03 eV phonon is gigantic (perhaps corresponding to the Cu-O bond bending mode [12]), the coupling with the background electronic continuum is not appreciable, since the phonon peaks are symmetric without showing Fano-like resonance, and the oscillator strengths do not change with x. The gigantic strength of the 0.03 eV mode is not specific to this system but a similarly strong phonon is commonly observed for other (non-Cu) transition-metal oxides with the K_2NiF_4 structure [12]. The oscillator strength may originate from the coupling with higher energy electronic excitations above 2 eV.

The low-energy electronic contributions to $\sigma_c(\omega)$ progressively increase and its ω dependence becomes appreciable for higher dopant concentrations. However, the values of optical conductivity are by orders of magnitude smaller than those of the in-plane conductivity in the corresponding energy range, and for the underdoped compounds even smaller than the Mott-Ioffe-Regel critical value $\sigma_c^{\min} \sim 50\ \Omega^{-1}\text{cm}^{-1}$ [13] which appears consistent with the nonmetallic transport in the c direction. $\sigma_c(\omega)$ for $x \leq 0.15$ is only weakly ω dependent, so that it cannot be described by a Drude formula even if an ω dependence of scattering rates is taken into consideration.

On the other hand, for the overdoped material with $x = 0.30$ the electronic contribution to $\sigma_c(\omega)$ allows a Drude fit to extract carrier scattering rate τ_c^{-1} as shown in Fig. 2(a). As the temperature is lowered, σ_c below ~ 0.03 eV is strongly enhanced, consistent with the T dependence of ρ_c, whereas σ_c in the higher energy region is only weakly T dependent—σ_c slightly decrease up to about 0.2 eV with lowering temperature so as to compensate the increased strength below ~ 0.03 eV. The result suggests that τ_c is determined by the larger scale between ω and T. The data actually fit the general expression $\tau_c^{-1} \sim \max(\omega^\alpha, T^\alpha)$ with $1 < \alpha < 2$, since 0.03 eV is comparable with the thermal energy at 300 K. The same ω- and T-dependent scattering is also suggested for the in-plane optical conductivity of the overdoped compound [14], and thus provides evidence for coherent charge transport in all three directions. The T dependence of $\sigma_c(\omega)$ is apparently similar for a superconducting compound with $x = 0.20$ [Fig. 2(b)] in conjunction with the positive T coefficient of ρ_c in this slightly overdoped material [9]. In this case it is possible to describe σ_c formally by the generalized Drude formula with $\tau_c^{-1} \sim \max(\omega, T)$ shown in the inset.

The Hall coefficient (R_H^c) measured under magnetic field ($\mathbf{B}$) applied parallel to the planes involves charge transport between the CuO_2 planes. However, there have been only a few attempts and reported results on $YBa_2Cu_3O_7$ are highly uncertain both in magnitude and in temperature dependence [15–18]. The present $La_{2-x}Sr_xCuO_4$ system has advantage in studying R_H^c, since large single crystals make it possible to investigate R_H^c in the two configurations, the current $\mathbf{J}$ parallel and perpendicular to the planes, and to minimize contact misalignment to which the Hall signal is very sensitive [18].

The experimental results are shown in Fig. 3 for three metallic compositions [also shown is the Hall coefficient in ordinary configuration ($\mathbf{B} \perp$ plane)]. For overdoped $x = 0.30$, R_H^c has small negative values ($\sim 10^{-4}$ cm^3/C, comparable with the results of the band calculation by Hamada *et al.* [19]) and is only weakly T dependent. The T dependence of the in-plane R_H^{ab} is also weak and tends

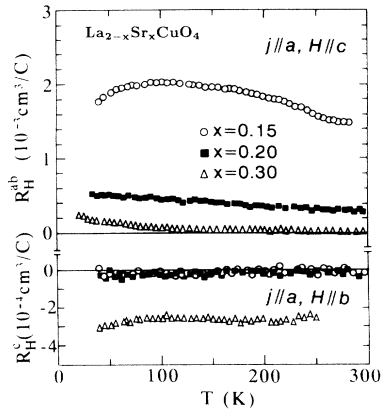

FIG. 3. Temperature dependence of the Hall coefficient for three metallic compositions of $La_{2-x}Sr_xCuO_4$ with $\mathbf{B} \perp \mathbf{c}$ (lower part) and with $\mathbf{B} \parallel \mathbf{c}$ (upper part).

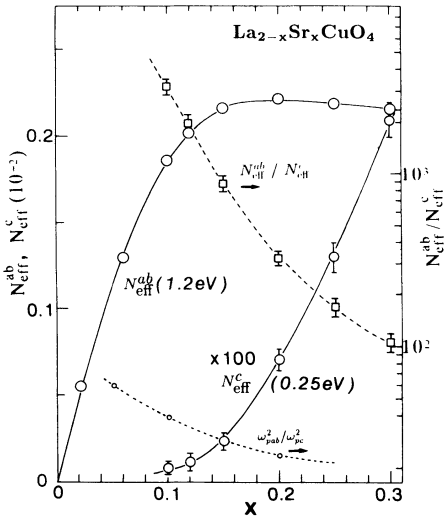

FIG. 4. Integrated spectral weight of in-plane ($N_{\rm eff}^{ab}$) and out-of-plane ($N_{\rm eff}^{c}$) electronic contribution plotted against x in $La_{2-x}Sr_xCuO_4$. The scale of $N_{\rm eff}^{c}$ is magnified by a factor 10^2. Also plotted is the anisotropic ratio $N_{\rm eff}^{ab}/N_{\rm eff}^{c}$ and corresponding values of $\omega_{pab}^2/\omega_{pc}^2$ ratio from the band theoretical calculation (Ref. [25]) on a logarithmic scale (the right-hand scale).

to be negative at higher temperatures. The Hall coefficients are therefore consistent with the metallic conduction in all directions. For superconducting compositions, on the other hand, $R_{\rm H}^{c}$ remains negative [20] but it becomes remarkably small in magnitude, while $R_{\rm H}^{ab}$ has large positive values and increases with reducing x. The contrasting behavior between $R_{\rm H}^{c}$ and $R_{\rm H}^{ab}$ is an additional evidence for different charge transport processes in the c direction which might arise from hopping due to random interplane scattering [21] or from quite other mechanisms including a exotic mechanism such as that in the resonating valence bond (RVB) state [22].

To discuss the x dependence of the electronic contribution to $\sigma_c(\omega)$, we examine the integrated spectral weight, $N_{\rm eff}^{c}(\omega) = (2mV/\pi e^2)\int_0^{\omega} \sigma_c^{(e)}(\omega')d\omega'$, where m and V are the bare electron mass and the unit cell volume, respectively, and $\sigma_c^{(e)}$ is obtained from $\sigma_c(\omega)$ by subtracting the optical phonon contributions. $(m_c/m)N_{\rm eff}^{c}(\omega)$ with m_c being the effective electron mass in the c direction is the effective number of electrons per unit cell contributing to $\sigma_c^{(e)}(\omega)$ below ω. The integrated $\sigma_c(\omega)$ including phonon contributions shows a jump at each energy corresponding to the optical phonons and the magnitude of each jump, the phonon oscillator strength, is independent of x. So, the phonon contributions can unambiguously be subtracted to yield purely electronic $N_{\rm eff}^{c}$. Since the low energy electronic conductivity becomes vanishingly small at ~ 0.25 eV for any x, $N_{\rm eff}^{c}(\omega)$ exhibits a plateau around this energy and thus $N_{\rm eff}^{c}$ at 0.25 eV (~ 2000 cm^{-1}) well approximates the total spectral weight of the low-energy electronic excitations.

We plot in Fig. 4 $N_{\rm eff}^{c}$ at 0.25 eV as a function of x together with an in-plane spectral weight $N_{\rm eff}^{ab}$ at 1.2 eV which represents the effective number of electrons involved in the low-energy excitations in the CuO_2 plane [1]. While $N_{\rm eff}^{ab}$ steeply increases upon doping and soon saturates for $x \geq 0.15$, $N_{\rm eff}^{c}$ remains very small and becomes appreciable only for $x > 0.15$. The variation of $N_{\rm eff}^{c}$ is strongly nonlinear with x ($N_{\rm eff}^{c} \sim x^{\alpha}$ with $\alpha \gtrsim 4$), and as a consequence, the anisotropic ratio $N_{\rm eff}^{ab}/N_{\rm eff}^{c}$ is extremely enhanced with decreasing x. For a comparison with the band theoretical calculation, we show in Fig. 4 the values of $\omega_{pab}^2/\omega_{pc}^2$, ω_{pab} and ω_{pc} being the in-plane and out-of-plane plasma frequency of conduction electrons, respectively, estimated from the result of the band calculation for $La_{2-x}Sr_xCuO_4$ [23]. The values of $\omega_{pab}^2/\omega_{pc}^2$ (~ 28 for $x = 0.15$) are by orders of magnitude smaller than the corresponding values of $N_{\rm eff}^{ab}/N_{\rm eff}^{c}$ [by definition ω_{pab}^2 (ω_{pc}^2) should be $4\pi e^2 N_{\rm eff}^{ab}$ ($4\pi e^2 N_{\rm eff}^{c}$)] and depend weakly on x [24].

Apparently, a substantial amount of the interplane spectral weight is missing in the lowest energy region (specifically below 0.25 eV) of the c-axis optical spectrum, which indicates that the charge dynamics is much more two-dimensional than anticipated by the band theory. Only for the overdoped compound ($x = 0.30$) the anisotropy tends to be comparable to the result of the band calculation and also to the resistivity anisotropy $\rho_c/\rho_{ab} \sim 100$ [9]. We should note that the rapid suppression of the c-axis spectral weight upon reducing hole density is not specific to the La214 system. Cooper *et al.* [25] reported basically the same behavior for $YBa_2Cu_3O_{6+x}$ with reduced oxygen content.

The observed semiconducting T dependence of ρ_c in the underdoped region and the very small $R_{\rm H}^{c}$ are apparently linked with the c-axis optical conductivity spectrum $\sigma_c(\omega)$ which is severely suppressed and nearly ω independent. The doping dependence of $\sigma_c(\omega)$ in high-T_c cuprates has recently been addressed from the two

contrasting theoretical viewpoints. One is based on the picture of a non-Fermi liquid [26]. According to the RVB scenarios with spin-charge separation, a spinon and a holon have to recombine to form a physical electron which hops between planes. Owing to the bosonic distribution of the holon momentum, the motion of the physical electrons between planes is strongly diffusive, so that the c-axis optical conductivity shows a broad continuum instead of a sharp peak at $\omega = 0$. The integrated spectral weight $N_{\rm eff}^{c}$ is then proportional to the holon density x (as well as the spinon density of states). In order to explain the observed strong x dependence of $N_{\rm eff}^{c}$, the theory has to presume, for example, a spin gap (a gap in the spinon density of states) [26,27], the presence of which has not been confirmed for the La214 system at the present moment [28].

The other viewpoint, based on a highly anisotropic Fermi liquid [29,30], incorporates interplanar disorder which assists the hopping between planes. In this case the interplane charge transport is determined by a competition between coherent Bloch wave propagation via the interplane hopping matrix element (t_c) and the interplane diffusion originating from random (incoherent) interplane scattering. The former process gives a Drude term and the latter does ω-independent background, and both processes are additive in σ_c. For $La_{2-x}Sr_xCuO_4$ the background term will overweigh the Drude term in the underdoped region and both terms will increase with x, as the interplanar disorder, as well as t_c due to the reduced Coulomb interaction, will increase. It should be checked whether this picture can explain the observed strong x dependence of $N_{\rm eff}^{c}$.

In summary, we have presented the x dependences of the c-axis optical spectrum and Hall coefficient of $La_{2-x}Sr_xCuO_4$ single crystals in the normal state. The results clearly demonstrate the change of the interplane charge dynamics with doping. The present experimental results will contribute to a deeper understanding of the electron states in the high-T_c cuprates.

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