In-plane optical conductivity of $La_{2-x}Sr_xCuO_4$: Reduced superconducting condensate and residual Drude-like response

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Temperature dependencies of the optical spectra of $La_{2-x}Sr_xCuO_4$ with x=0.12 and 0.15 were carefully examined for a polarization parallel to the CuO_2 -plane over a wide frequency range down to 8 cm⁻¹. Selection of well-characterized crystals enabled us to measure purely in-plane polarized spectra without any additional peak. The weight of superconducting (SC) condensate estimated from the missing area in $\sigma_1(\omega)$ well agrees with the estimate from the slope of $\sigma_2(\omega)$ vs $1/\omega$ plot, showing no evidence that the Ferrell-Glover-Tinkham sum-rule is violated in the optical spectrum. We demonstrate that the optically estimated SC condensate is much smaller than the value obtained from the μ SR measurement of magnetic penetration depth. We also find an anomalous increase of conductivity in submillimeter region towards $\omega=0$ below T_c , which suggests the microscopic inhomogeneity in the superconducting state. Both observations are discussed in relation with the inhomogeneous electronic state that might be inherent to high- T_c cuprates.

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I. INTRODUCTION

The superconducting (SC) condensate, one of the essential parameters for a superconductor, has been a subject of discussion in high- T_c superconducting cuprates (HTSC). The recent progress in scanning tunneling spectroscopy has revealed a strange inhomogeneous distribution of the gap in Bi₂Sr₂CaCu₂O₈ (BSCCO).¹ The effect of this real-space gap inhomogeneity on a superconducting (SC) condensate is unclear. On the other hand, the angle-resolved photoemission spectroscopy (ARPES) has demonstrated a distinct *k*-dependence of the gap at the Fermi surface in the pseudogap state or the stripe ordered state.^{2,3} For the system with such a partially gapped Fermi surface, the source of SC condensate is not obvious.

As one of the direct methods to detect a SC gap and the weight of SC condensate, optical studies have been extensively performed for HTSC. A clear conductivity depression was observed below ~500 cm⁻¹ in YBa₂Cu₃O_y (YBCO) (Refs. 4–7) and BSCCO.⁸ Then, the weight of the SC condensate ($\rho_s \sim \omega_{ps}^2$) was estimated from the missing area (A) of the real part $\sigma_1(\omega)$ of conductivity spectrum or from the low-frequency behavior of its imaginary part $\sigma_2(\omega)$.

The estimate of SC condensate from optical spectra, however, has been subject to uncertainty. One of the reasons is that, unlike the case of conventional superconductors, there often remains some conductivity below the gap energy, forming a Drude-like increase towards $\omega = 0.^9$ Since the missing area of $\sigma_1(\omega)$ is large in YBCO and BSCCO, the remaining conductivity effect is relatively small in estimation of SC condensate. By contrast, in the case of lower T_c -HTSC such as La_{2-x}Sr_xCuO₄ (LSCO), it may affect the result more seriously. Nevertheless, this residual conductivity was ignored in most of the previous estimate of SC condensate except for a few works.^{10,11} The origin of this remaining conductivity is an important problem to be solved.

The other reason for the difficulty in determining SC condensate of LSCO is linked to the difficulty in obtaining a pure in-plane spectrum. Since the early stage of HTSC research, various different in-plane spectra have been reported for LSCO (Refs. 10–15) and $La_2CuO_{4+\delta}$ ^{16,17} Some of the reported in-plane spectra show rich features in a far-infrared (FIR) region,^{13,14,16} which are ascribed either to the LO-phonons¹³ or to the excitation related to the polarons.¹⁴ According to the variety of spectra, different values of the London penetration depth $\lambda_L^{\text{FIR}}(=1/2\pi\omega_{ps})$ were derived, ranging from 250 to 430 nm for the samples near x=0.15, 10-14,16 which do not correlate with the T_c -value of the studied sample. Namely the scattered data cannot be explained by the possible difference in Sr-content or oxygen content. Some of them are considerably different from the value determined by muon spin rotation (μSR) measurements.18,19

The purpose of this work is twofold. The first one is to clarify the origin of such a confusing situation in optical study of LSCO. We report a purely in-plane polarized spectrum of LSCO and demonstrate the effect of the c-axis component mixing, by comparing with the "dirty" spectra contaminated by the *c*-axis component. Possible reasons for the c-axis component mixing are discussed. The second and main purpose is to examine the electrodynamic response in the low energy region and to estimate the SC condensate, based on the purely in-plane polarized optical spectra of $La_{2-x}Sr_xCuO_4$ (x=0.12 and 0.15). The spectra were measured over a wide range of frequency 8-30000 cm⁻¹, down to the submillimeter region. At $T \ll T_c$, we have found a depression of conductivity below 200 cm⁻¹ (70 cm⁻¹) for x=0.15 (x=0.12) but a steep increase of the conductivity below 40 cm^{-1} (10 cm^{-1}), which leads to a significantly small missing area compared to the value estimated from the London length λ_L determined by μ SR measurement. We conclude that a discrepancy of the SC condensate between

the optical and μ SR-estimations is commonly observed in many HTSC. Possible scenarios for these anomalous features are discussed.

II. EXPERIMENTS

Single crystals of $La_{2-x}Sr_xCuO_4$ with x=0.12 and 0.15 were grown by a traveling solvent-floating zone (TSFZ) method. The superconducting transition temperature T_c is 36 K for x=0.15 and 30 K for x=0.12. Fixing crystal axes by observing x-ray Laue patterns at the surface of as-grown crystal rods, we cut the crystals along the *c*-axis and took several pieces of samples with a thickness of about 2 mm. We call them "*ac*-face" sample hereafter. For comparison, we also cut some crystal pieces with the *c*-axis perpendicular to the measurement surfaces called "*ab*-face" sample. In both cases, *a*- and *b*-axes are not distinguished although there is a small difference between them. After a number of trial measurements of far-infrared spectra, we realized that even Laue-characterized crystals were sometimes multidomains, showing the *c*-axis phonon admixture.

The second characterization method to detect a multidomain is to observe the surface using a polarized optical microscope. Prior to this observation, the sample surfaces were polished with Al_2O_3 powder of different particle sizes at several steps. The particle diameter for the final polishing was 0.3 μ m. It was found that some pieces of crystals consisted of multidomains. We carefully selected the single domain crystals for measurement of a purely in-plane polarized spectrum. A typical diameter of the sample disks is about 5 mm. A spot size of the incident light is about 3 mm, smaller than the sample size.

Optical reflectivity spectra were measured using a coherent-source spectrometer²⁰ in a millimeter and submillimeter wavelength region $(8-33 \text{ cm}^{-1})$, a Fourier transformation type spectrometer for infrared region $(30-8000 \text{ cm}^{-1})$, and a grating type one for higher energy region above nearinfrared $(4000-30000 \text{ cm}^{-1})$. Both a sample and a gold mirror were mounted on a copper plate in a He-gas flow cryostat for temperature control. Before each measurement of a sample spectrum, a spectrum of the gold mirror was measured as a reference. Since high accuracy of reflectivity value is required in a millimeter and submillimeter region where reflectivity approaches 100%, we took the literature values of gold reflectivity instead of assuming 100% reflectivity of gold to calculate the absolute values of sample reflectivity. Reflectance of polarized light was measured with the s-polarized geometry at the incident angle of about 5°.

In order to compare the SC condensate estimated from the other experimental methods, we measured the London penetration depth using muon spin rotation (μ SR) technique. To improve the reliability of the missing area analysis and discussion, the μ SR measurement has been carried out on the same single crystal as used in the optical study. The analysis of the field distribution of a vortex lattice in the applied transverse field H_i =0.2 Tesla has been performed using a London model²¹ with a Lorentzian cut-off²² of 4.5–6.5 nm as a nonlocal correction. Other possible contributions for the broadening of the field distribution, such as random pinning,



FIG. 1. Comparison of the in-plane reflectivity spectra of two $La_{1.85}Sr_{0.15}CuO_4$ crystals at 300 K. Both of LSC#1 and LSC#2 were measured on the *ac*-faces with polarization $E \parallel a$. The spectra of LSC#2 shows a substantial contribution from the *c*-axis spectrum. The *c*-axis spectrum of LSCO is indicated by a gray curve as a reference. We can reproduce a similar profile to LSC#2, using the pure in-plane spectrum (LSC#1) and the *c*-axis spectrum, as described in the text. The calculated reflectivity assuming a 15° rotation around the *ac*-/*bc*-plane or equivalently the 6.7%-admixture of the *c*-axis component is indicated by a short-dashed curve.

are corrected by assuming an additional Gaussian relaxation with the relaxation rate 0.39–0.43 MHz.

III. RESULTS AND ANALYSIS

A. Effect of *c*-axis component admixture

Figure 1 shows the far-infrared reflectivity spectra at room temperature for several samples. LSC#1 is the almost perfectly in-plane polarized spectrum measured for the *ac*-face of the La_{1.85}Sr_{0.15}CuO₄ crystal, while LSC#2, the spectrum for another *ac*-face La_{1.85}Sr_{0.15}CuO₄ crystal cut from different part of the rod, shows the bumps centered at around 300 cm⁻¹ and 520 cm⁻¹. It turns out that the frequencies of these structures coincide with those observed as phonon reststrahlen bands in the reflectivity spectrum with *c*-polarization (*E*||*c*). Namely, the bumps clearly result from mixing of the *c*-polarized component into the in-plane spectrum.

We can reproduce a spectrum similar to that of LSC#2, using LSC#1 as a pure in-plane reflectivity data $(R_{a/b})$ and the measured *c*-polarized reflectivity data (R_c) , with the simple formula²³ $R_{\theta} = R_{a/b} \cos^2(\theta) + R_c \sin^2(\theta)$. The shortdashed curve in Fig. 1(a) represents the calculated reflectivity spectrum for $\theta = 15^{\circ}$ deviation of the polarization direction from the pure a(b)-axis or equivalently the 6.7%admixture of misoriented volume. Corresponding to the rapid drop of reflectivity above 600 cm⁻¹, the dip at ω $\sim 470 \text{ cm}^{-1}$ and the suppression below 250 cm⁻¹ in the *c*-axis spectrum, the weak reflectivity suppressions giving the bump features in LSC#2 are well reproduced in the calculated spectrum. A good demonstration was made in the optical spectra of intentionally misaligned films of $(La, Ce)_2CuO_4$ ²⁴ where only 1° misorientation results in a clear feature of the *c*-axis phonons in the spectra.



FIG. 2. (a) Comparison of the in-plane conductivity $\sigma_1(\omega)$ of La_{1.85}Sr_{0.15}CuO₄ (LSC#1) and the *c*-axis component mixed data for θ =15°. (b) Comparison of the scattering rate $1/\tau(\omega)$ for LSC#1 and the *c*-axis component admixture case. It is clear that the double peak is a result of the *c*-component admixture. It is also demonstrated that the kink at ~700 cm⁻¹ even at 300 K and is independent of the superconducting response below 200 cm⁻¹.

Other than the exposure of the *c*-axis optical phonons in the "nominal" in-plane spectrum, the admixture of the *c*-axis spectral component seriously influences the conductivity and scattering rate spectra calculated from the reflectivity spectrum via Kramers–Kronig (KK) transformation. As shown in Fig. 2(a), the *c*-component admixture creates a broad conductivity dip between 300 and 1000 cm⁻¹, a sharp peak around 500 cm⁻¹ and a broad peak below ~ 200 cm⁻¹. These are artifacts arising from *c*-axis admixing: The low- ω broad peak arises as a consequence of suppression of reflectivity in the lowest frequency region due to the superposition of the "insulating *c*-axis spectrum." Its peak height and position are quite sensitive to a small change in magnitude of the low- ω reflectivity as well as a change in the $\omega \rightarrow 0$ extrapolation of the spectrum. These features were identified in the reported spectra of LSCO (Refs. 13 and 14) and La₂CuO_{4+ δ}¹⁶ which we interpret to indicate the admixture of the *c*-axis component. Figure 2(b) demonstrates that the double peak in $1/\tau(\omega)$ reported by Startseva *et al.*¹³ can also be considered as the same admixture effect.

There are several possible causes for the *c*-axis component mixing, such as (i) miscut of crystals, (ii) multidomains in as-grown crystals, (iii) *p*-polarization admixture. Miscut of as-grown crystals is the most probable mistake that gives a misoriented surface for reflectivity measurement. This is the case particularly with the LSCO crystals, because the cutting procedure is inevitable for extracting a sizable sample from an as-grown crystal rod. The effect of miscut is serious in measurement on the *ab*-face, while the measurement on the *ac*-face is not affected by the miscut as far as it is done with polarization perpendicular to the *c*-axis and per-

pendicular to the incident plane (*s*-polarization). In the case of YBCO and BSCCO crystals grown by a flux method, natural surfaces can be used for optical measurement. The BSCCO crystals, even if grown by the TSFZ-method, are so easily cleaved that crystal cutting is not necessary to extract the *ab*-face samples.

The multidomain problem is serious in the TSFZ-grown crystals of LSCO and the related compounds. It often appears in the early stage of crystal growth, namely, in the crystal portion close to a seed. It is hard to confirm that a sample is of single domain by a usual x-ray Laue pattern measurement, because a typical x-ray spot size is much smaller than the sample surface area of about 3×3 mm². This was the case for LSC#2. In order to scan the whole area of sample surface, we need an x-ray topography measurement. An easier way to examine the existence of a multidomain feature is to observe a polished sample surface using a polarized optical microscope. Even if a sample passes these checks, there could be cases where the far-infrared spectrum shows some admixture of different direction component. The misoriented domain hidden behind the surface within a penetration depth (~1 μ m) of far-infrared light is a possible origin. Although the dielectric response of the light penetrating through multidomains would be generally complicated, and the obtained reflectivity could not merely be expressed by the weighted summation R_{θ} , the spectrum with the *c*-axis component admixture shows the artificial features addressed above.

Even when we have samples with pure *ab*-plane surface, a problem related to the *p*-polarization must be considered in the case of measurement without using a polarizer. In this case, the *c*-axis component manifests itself as notches in the in-plane reflectivity spectrum at the frequencies of LOmodes, as was demonstrated by Van der Marel et al.25 to explain the spectra reported by Reedyk and Timusk.²⁶ Van der Marel et al. pointed out that in a typical reflectivity measurement with the incident angle of 8°-12° the electric field parallel to the plane of incidence (*p*-polarization) causes the c-axis phonon modes to leak. In order to gain a signal, we sometimes did not use a polarizer for the *ab*-face measurement because the light intensity is reduced to about a half when going through a polarizer. Since unpolarized light should have a component of *p*-polarization, this type of measurement has a possibility to yield a spectrum with features corresponding to the *c*-axis phonons. Such possibility is seen in part of the data by Lucarelli et al.¹⁴ About a half of the seven spectra were measured on the ab-face without a polarizer, and they show a clear feature corresponding to the c-axis phonon.²⁷

All the problems due to the *c*-axis component mixing become serious only in strongly anisotropic systems, namely, when the reflectivity values are markedly different in the *ab*and *c*-polarized spectra. In fact, less anisotropic materials such as well-oxygenated YBCO with metallic *c*-axis conduction have seldom shown the *c*-axis component in the spectrum measured with the in-plane polarized light. Therefore, the measurement of in-plane spectrum for LSCO and probably TFSZ-grown (Nd, Ce)₂CuO₄ requires particularly high quality samples without any multidomain feature, high accuracy in cutting angle, and careful polarization measurements.



FIG. 3. (Color online) Temperature dependence of in-plane reflectivity spectra of $La_{2-x}Sr_xCuO_4$ with x=0.15 and x=0.12 with E||a| or b. The inset of the upper panel shows the spectra for x = 0.15 in submillimeter (SMM) and far-infrared (FIR) regions separately. The inset of the lower panel shows the room temperature spectra in a wider range of frequency.

(The measurement on the *ac*-face with *s*-polarization geometry is most preferable to avoid the admixture of the *c*-axis component.) These requirements are quite severe, compared to that for other measurement techniques such as neutron scattering.

B. In-plane optical spectra

Hereafter we focus on the results of the sample for LSC#1 which we believe the purely in-plane polarized spectrum, and for comparison on the results of the x=0.12 sample with the *ac*-face which was also well characterized. Figure 3 shows the in-plane reflectivity spectra for x=0.15 (a) and



FIG. 4. (Color online) Temperature dependence of in-plane conductivity spectra of $La_{2-x}Sr_xCuO_4$ with x=0.15 and x=0.12 with $E \parallel a$ or *b*. Inset figures show the low- ω spectra and the dc conductivity. The optical data can be smoothly extrapolated to the dc values.

0.12 (b) at various temperatures. Phonon peaks are almost screened by free carriers except for the small structure at ~370 cm⁻¹ ascribed to the in-plane oxygen bending mode phonon. No admixture of the *c*-axis component is observed in both sample spectra. The FIR reflectance is smoothly connected to the sub-millimeter reflectance as shown in the inset of Fig. 3(a). When the temperature is lowered below T_c , the reflectance for x=0.15 exhibits a rapid increase below 150 cm⁻¹ and a plateau below 40 cm⁻¹, which could be originating from the transfer of low frequency spectral weight to $\delta(\omega)$ at $\omega=0$. A similar feature is observed in the in-plane SC spectra of YBCO and BSCCO, but at higher frequency of ~500 cm⁻¹, probably owing to the larger energy scale of SC gaps.

The reflectance is transformed to the complex conductivity $\sigma(\omega)$ by utilizing KK relations. Figure 4 shows the real part of the conductivity $\sigma_1(\omega)$ that can be smoothly extrapolated to a dc value at each temperature above T_c as shown in the inset, justifying the validity of our KK analysis. The low- $\omega \sigma_1(\omega)$ for x=0.15 increases with decreasing temperature, forming a Drude-like peak at $\omega = 0$, as is seen in Fig. 4(a). When the temperature decreases below T_c , σ_1 starts to be suppressed below 250 cm⁻¹, corresponding to the SC gap opening. This SC response is, however, followed by an unusual Drude-like increase below 40 cm⁻¹ even at 5 K \ll T_c. A similar Drude-like response is also seen in the low- ω upturn of the spectrum for x=0.12 [Fig. 4(b)]. The Drude-like peak width is narrower in the x=0.12 sample than that for x =0.15. A huge amount of residual conductivity at $T \ll T_c$ has also been reported in a direct measurement of conductivity of $La_{1.84}Sr_{0.16}CuO_4$ film in the submillimeter wavelength region.¹⁰ Moreover, this feature seems to be common in many HTSC such as YBCO and BSCCO,⁹ which should affect the estimation of the SC condensate.

To compare with the previous results of LSCO,¹³ YBCO,²⁸ and BSCCO,²⁹ we derive an ω -dependent scattering rate $1/\tau(\omega) = (\omega_p^2/4\pi) \operatorname{Re}[1/\sigma(\omega)]$ for x=0.15 as plotted in Fig. 2(b). Here ω_p is obtained from the integral $\int_0^{\omega_0} \sigma_1(\omega) d\omega = \omega_p^2/8$. We take $\omega_0 = 10000 \text{ cm}^{-1}$ so that the integral range includes the reflectance plasma edge, but is well below the charge transfer excitation energy ($\sim 16000 \text{ cm}^{-1}$) in La₂CuO₄.³⁰ The spectrum of $1/\tau(\omega)$ clearly shows a kink at 650 cm⁻¹ at all temperatures. The kink in $1/\tau(\omega)$ is commonly observed at nearly the same frequency at low temperatures for many HTSC studied so far.²⁹ This has been ascribed to the opening of a pseudogap (or a SC gap) and/or the coupling of carriers with a magnetic collective mode observed as a resonance peak in neutron scattering.³¹ An alternative candidate for this kink is the coupling with a phonon that is proposed as a source for the kink in energy-dispersion curve of angle-resolved photoemission spectrum (ARPES).³²

The present result confirms that $1/\tau(\omega)$ shows a kink at 700-800 cm⁻¹ as many other HTSCs do. The $1/\tau$ -kink in LSCO is different from that in YBCO or BSCCO in that it is not a low-T phenomenon but is observable at room temperature or even higher for an optimally doped sample (x=0.15). It is not clear whether the kink is caused by the pseudogap opening, because there is a controversy on the presence or otherwise of the pseudogap in LSCO. From the T-dependence of in-plane resistivity ρ_{ab} , a pseudogap temperature is claimed to be as low as 70–80 K for x=0.15,³³ whereas the temperature below which the uniform susceptibility starts to decrease (or the Hall coefficient starts to increase³⁴) is higher than 300 K. What is clear with the kink in LSCO is that it is not directly linked to T^* in the $\rho_{ab}(T)$ -curve. Another important result is the suppression of $1/\tau$ upon opening of the SC gap is observed at $\omega \sim 100 \text{ cm}^{-1}$ that is far below the kink frequency. This is in sharp contrast to the continuous development from a pseudogap to a SC gap observed in the other HTSC such as YBCO and BSCCO.²⁸ The present observation suggests that the kink feature has nothing to do with a SC gap, and probably is not related to pseudogap. We think that the kink in $1/\tau$ represents a crossover from the low- ω charge dynamics to the high- ω one. The former as well as the dc conductivity is governed by the Fermi surface around the nodal point, while the electronic excitations in the antinodal region are also involved in the latter.

IV. DISCUSSIONS

A. SC condensate

The observed SC response is most naturally interpreted as the signature of formation of SC condensate associated with a SC gap opening. When a delta-function peak develops at $\omega=0$ in $\sigma_1(\omega)$ at the expense of its suppression in the low- ω region, $\sigma_2(\omega)$ behaves as proportional to ω_{ps}^2/ω . There are two ways to estimate the SC condensate ω_{ps}^2 from the optical



FIG. 5. (a) Optical conductivity of La_{1.85}Sr_{0.15}CuO₄ (T_c = 36 K) with $E \parallel a$ or *b* at (T=40 K) and 5 K. The missing area $A_{\mu SR}$ expected from λ_L =280 nm obtained in the μ SR study is shown by a hatched area. The solid circle indicates the dc conductivity at 40 K. (b) The ratio of the missing area obtained from the present optical study $A(\omega_0)$ to $A_{\mu SR}$. MA#1 represents the ratio for $A(\omega_0)$ calculated with the lower limit of integral of 8 cm⁻¹. MA#2 is for the data of $A(\omega_0)$ calculated by the dashed curve extrapolation in (a).

spectrum. One is to calculate the missing area $A(=\omega_{ps}^2/8)$ in $\sigma_1(\omega)$, and the other is to estimate ω_{ps} from the slope of $\sigma_2(\omega)$ vs $1/\omega$. In the case of HTSC, it is not a trivial issue that these two methods give an identical value of ω_{ps} , since a possibility of kinetic energy contribution has been suggested by both theory³⁵ and experiment.^{36,37} We compare the two optically obtained values to check the Ferrell-Glover-Tinkham (FGT) sum rule, and also compare them with those estimated from the London penetration depths that were determined by μ SR measurement.

Below, we focus on the condensate weight at the lowest temperature (~5 K) well below T_c . The conductivity missing area for x=0.15 is defined by the integral

$$A(\omega_0) = (120/\pi) \int_{0+}^{\omega_0} [\sigma_1(\omega, 40 \text{ K}) - \sigma_1(\omega, 5 \text{ K})] d\omega, \quad (1)$$

where we set $\omega_0 = 10000 \text{ cm}^{-1}$. We first put the lower limit of the integral at 8 cm⁻¹ which is the limit of our optical measurement (labeled as MA#1). In Fig. 5(a), the missing area $A_{\mu\text{SR}} = 1/8(2\pi\lambda_L^{\mu\text{SR}})^2$ expected from $\lambda_L^{\mu\text{SR}} = 280 \text{ nm}$ obtained by the μ SR study³⁸ is shown by the hatched area. It is obvious that $A_{\mu\text{SR}}$ is much larger than the missing area in the optical conductivity spectrum between 40 and 5 K. The ratio of the optical missing area $A(\omega_0)$ and the μ SR value $A_{\mu\text{SR}}$ is plotted in Fig. 5(b) as a function of ω_0 . $A(\omega_0)/A_{\mu\text{SR}}$ increases and then saturates to 0.25 ± 0.05 , showing no appreciable change up to 10000 cm⁻¹. The penetration depth corresponding to this value of $A(\omega_0)$ is 530 nm. Even if we eliminate the contribution of the Drude-like component at 5 K and replace it by a straight line [shown by a dashed line in Fig. 5(a)] which mimics a *d*-wave SC behavior, $A(\omega_0)/A_{\mu SR}$ amounts only to 0.5. This is plotted as a dashed curve labeled as MA#2 in Fig. 5(b), giving an upper limit of the missing area (or the lower limit of λ_L^{FIR} =400 nm) in our experiment.

The anomalously small missing area is also observed for x=0.12. Compared to the SC condensate estimated from the μ SR λ_L^{μ SR}=310 nm, the optical conductivity missing area $A(\omega_0)$ is substantially small with the ratio $A(\omega_0 = 10000 \text{ cm}^{-1})/A_{\mu}$ SR ~ 0.1 . The optically determined penetration depth turns out to be $\sim 1 \mu$ m in this case.

Strictly speaking, the missing area which is connected with the SC condensate should be calculated from the difference between the normal and the superconducting conductivity at T=5 K. In the above estimation of $A_0(\omega_0)$, we assume that $\int \sigma_1(\omega, 40 \text{ K}) d\omega = \int \sigma_1^n(\omega, 5 \text{ K}) d\omega$, where $\sigma_1^n(\omega, 5 \text{ K})$ is the conductivity in the case that the system keeps the normal state down to T=5 K. In a conventional metal (superconductor), this assumption holds well, because the low-T resistivity is almost constant (the residual resistivity regime). In the case of LSCO with x=0.12 and 0.15, the "normal state" resistivity realized by the application of intense magnetic fields increases weakly with lowering T below T_c ,³⁹ suggestive of a "normal" state rather similar to the conventional one. As regards the observed T-dependence of the normalstate spectrum, Fig. 4 (and its inset) indicates that the frequency range where σ_1 shows an appreciable T-variation shrinks with decreasing T. For the spectrum at T=50 K, a difference from that at T=100 K is seen restricted below 30 cm⁻¹. The trend suggests that the T-dependence of σ_1^n arises predominantly from the Drude term. From this, and also from the result of dc resistivity measured under intense fields, we guess that, if the normal state persists below T_c , the Drude term would not change significantly from that at T_c , and hence the difference between $\sigma_1(40 \text{ K})$ and $\sigma_1^n(5 \text{ K})$ would be small.

Another problem in estimation using Eq. (1) is the possible contribution from the higher frequency spectrum. Recently the spectral weight reduction in the high energy region above 10000 cm⁻¹ has been observed in BSCCO by two independent methods and the change in kinetic energy at superconducting transition has been reported.^{36,37} We have also found small *T*-dependence of the present spectra in the visible frequency region, which gives additional weight to $A(\omega_0=10000 \text{ cm}^{-1})$. However, the integral up to $\omega_0 = 30000 \text{ cm}^{-1}$ increases $A(\omega_0)$ only by 20% at most. Possible kinetic energy contribution to the condensation energy cannot explain the small missing area in the present results.

The second method to estimate the SC condensate is to use σ_2 . In Fig. 6, σ_2 is plotted as a function of $1/\omega$. It is substantially deviated from the linear-relationship, because a large amount of remaining conductivity $\sigma_1(\omega)$ below 50 cm⁻¹ strongly influences $\sigma_2(\omega)$. In order to remove the contribution of this residual conductivity, we adopt the method proposed by Dordevic *et al.*⁴⁰ First, we assume $\sigma_1(\omega)$ at 5 K as the conductivity of unpaired carriers $\sigma_1^{\text{res}}(\omega)$ which do not condense at $\omega=0$. Next, we calculate $\sigma_2^{\text{res}}(\omega)$



FIG. 6. Imaginary part of condutivity $\sigma_2(\omega)$ of $\text{La}_{1-x}\text{Sr}_x\text{CuO}_4$ for x=0.12 and 0.15 at low frequencies. $E \parallel a(b)$ and T=5 K. σ_2^{res} is calculated from the residual $\sigma_1(\omega)$ at 5 K using the KK-relation. When σ_2^{res} is substracted, σ_2 scales well with $1/\omega$, which is the signature of the superconducting carrier response.

corresponding to $\sigma_1^{\text{res}}(\omega)$ using a KK-relation. Then, the $\sigma_2^s(\omega)$ which should represent the "true" superfluid response is obtained as $\sigma_2^s(\omega) = \sigma_2(\omega) - \sigma_2^{\text{res}}(\omega)$.

The obtained $\sigma_2^s(\omega)$ is proportional to $1/\omega$ both for x =0.12 and 0.15, indicating a SC response in this frequency range. The SC condensates giving $\lambda_L^{FIR} = 990$ nm for x =0.12 and 590 nm for x=0.15 are estimated from the slopes, respectively. These are in agreement with the values (1 μ m for x=0.12 and 530 nm for x=0.15) obtained from the missing area in $\sigma_1(\omega)$. It means that this method for estimation of SC condensate is self-consistent within the optical spectra, and thus it is concluded that the FGT sum rule almost holds within an infrared frequency range. The same conclusion has been drawn for optimally doped YBCO in the recent paper by Homes et al.⁴¹ The present result does not completely exclude a possibility of small contribution from the high- ω region, namely, the kinetic energy contribution to the condensate weight, as was observed in BSCCO,^{36,37} but it is beyond the accuracy of our measurement.⁴²

The previously reported values of λ_L^{FIR} for the optimally doped LSCO range from 250 nm to 430 nm, depending on the sample and the research group but not on the T_c -value. From the simulation, as in Figs. 1 and 2, we learned that the value of λ_L^{FIR} estimated from the *c*-axis admixed spectrum is strongly dependent on how and to which extent the *c*-component is mixed, as well as the accuracy of the low- ω reflectivity measurement. In some case, as we demonstrated in Fig. 2(a), the admixture of the *c*-axis component tends to form an artificial peak at low frequency in the normal-state conductivity, which is not robust against a small change in the magnitude and $\omega \rightarrow 0$ extrapolation of reflectivity. Such an artificial peak in the normal-state conductivity leads to a large error in estimation of missing area.

However, the large scattering in the value of λ_L^{FIR} in the previous works mainly arises from whether or not the low- ω residual spectral weight in the SC state is correctly accounted. If the measurement is not done to low enough frequencies and the low- ω spectrum is assumed to be flat with R=1, the missing area in $\sigma_1(\omega)$ must be overestimated. Since the residual σ_1 strongly affects the low- ω behavior of σ_2 , the estimation of λ_L^{FIR} from σ_2 is also subject to large uncertainty, unless the residual "normal-state" component is sub-

tracted from σ_2 . The present results are free from these problems. The spectra were measured down to 8 cm⁻¹ and the residual conductivity was taken into account for evaluation of SC condensate within the error shown in Fig. 5.

The relatively larger missing area (or equivalently the shorter $\lambda_L^{\text{FIR}} = 250 \text{ nm}$), reported in the paper by Gao¹² may partly result from this overestimation of the missing area, because the 5 K- $\sigma_1(\omega)$ was set to zero below 70 cm⁻¹. A similar problem is seen in the result by Quijada¹⁶ reporting $\lambda_L^{\text{FIR}} = 310 \text{ nm.}$ Although the low- ω residual conductivity was clearly detected by Startseva,¹³ it was not subtracted when they estimated λ_L^{FIR} from the plot of ϵ_1 vs ω^{-2} , resulting in an overestimation of SC condensate. The measurement by Gorshunov *et al.*¹⁰ on the La_{1.84}Sr_{0.16}CuO₄ film (T_c =39.5 K),⁴³ which covered the frequency range down to 5 cm^{-1} , clearly detected a narrow Drude-like band with the large weight $\omega_p \sim 7800 \text{ cm}^{-1}$ at 5 K. Subtracting the Drude-like spectral contribution by fitting, they obtained a long penetration depth λ_L^{FIR} =400 nm. A little longer penetration depth λ_L^{FIR} =430 nm was reported by Somal *et al.*¹¹ for a single crystal. These values are in fairly good agreement with our estimate of the shorter limit of λ_L^{FIR} . Therefore, we conclude that the spectra successfully measured give a similar value of λ_L^{FIR} (400-500 nm) that is longer than the estimate (250-310 nm) for which the low- ω residual conductivity is not correctly evaluated.^{12,13,16}

The microwave (MW) conductivity measurement gives a similar large value $(=\lambda_L^{MW}=400\pm100 \text{ nm} \text{ for } x=0.15)$ to our FIR result, although the error bar is quite large in this technique.⁴⁴ By contrast, the μ SR penetration depth^{18,19} is clearly shorter than the FIR and microwave values. For x = 0.12, our estimate from the FIR data is $\lambda_L^{FIR} = 990 \text{ nm}$, the recent FIR data for x=0.125 by Dumm *et al.*⁴⁵ shows $\lambda_L^{FIR} = 630 \text{ nm}$, and the microwave data for x=0.12 is $\lambda_L^{MW} = 500\pm200 \text{ nm}.^{44}$ Note that λ_L^{FIR} as well as T_c is very sensitive to the Sr-content x around x=0.12 owing to the so-called 1/8-anomaly⁴⁶ (T_c is most suppressed when x is tuned to 0.115). Even if taking into account this situation, we can conclude that the values of λ_L^{FIR} are much longer than the μ SR estimate $\lambda_L^{\mu SR} = 310 \text{ nm}.$

A similar discrepancy between the FIR- and the μ SR-penetration depths is also seen in the case of YBCO and BSCCO particularly in the underdoped regime. In the optimally doped YBCO, the average value of a- and *b*-polarized data ($\lambda_L^{\text{FIR}} = 160 \text{ nm}$ for $E \parallel a$ and 117 nm for $E \parallel b$) (Refs. 6 and 7) is larger than the μ SR value $[\lambda_L^{\mu SR}(ab) = 112 \text{ nm} (\text{Ref. 47})]$. Comparing the conductivity spectra of optimally and underdoped YBCO,28,48 we find that the missing area of σ_1 in the underdoped YBCO with T_c =56-59 K is about 20% of that for the optimally doped one with $T_c = 93$ K. This suggests the penetration depth of about 300-400 nm for the underdoped YBCO. The recent estimation of λ_L^{FIR} by subtracting the low- ω residual conductivity is about 280 nm for YBa₂Cu₃O_{6.6}.⁴¹ This is much longer than the μ SR value (λ_L^{μ SR} = 170 nm).⁴⁷ A longer FIR penetration depth is also reported for underdoped BSCCO with T_c = 70 K.³⁷ Compared with the μ SR penetration depth (~190 nm) for BSCCO with $T_c = 75$ K,¹⁹ the reported value of λ_1^{FIR} (=680 nm) for the T_c =70 K sample³⁷ is extremely

TABLE I. Comparison of penetration depths estimated from FIR and μ SR measurements. (The FIR data of YBa₂Cu₃O_y are for $E \parallel a$, while the μ SR data are the average within the *ab*-plane.)

	$La_{2-x}Sr_xCuO_4$		YBa ₂ Cu ₃ O _y		Bi ₂ Sr ₂ CaCu ₂ O _z
	x=0.15	<i>x</i> =0.12	$y \approx 6.9$	$y \approx 6.6$	$T_c \approx 70 \text{ K}$
λ_L^{FIR} (nm) $\lambda_L^{\mu \text{SR}}$ (nm)	400–530 280 ^e	630 ^a –990 310 ^f	160 ^b 112 ^g	280 ^c 170 ^g	680 ^d 190 ^f
From Ref. 45.					

^bFrom Ref. 7.

^cFrom Ref. 41.

^dFrom Ref. 37.

^eFrom Ref. 38.

^fFrom Ref. 19.

^gFrom Ref. 47.

long. We summarize the comparison of the FIR- and μ SR-penetration depths for various HTSC in Table I. As is clearly seen in the table, the discrepancy between the FIR and μ SR data is quite robust for HTSC.

One may consider the possibility that a FIR spectrum is not correctly measured by some reason, for example, by the [110] surface geometry problem arising from the *d*-wave symmetric gap, as recently pointed out by Tu *et al.*⁴⁹ It is, however, unlikely because a small estimate of SC condensate was obtained by various measurements with various surface geometries.

It is also a general trend that the discrepancy becomes larger as one goes to more underdoped regime. This is suggestive of electronic inhomogeneity as a possible origin of the discrepancy. The SC order parameter may not be uniform in real space and vanish in some parts, according to the recent observation of STM on BSCCO.¹ In LSCO, the stripe fluctuation may introduce electronic inhomogeneity. As we discuss below, the presence of nonsuperconducting area appears to be correlated with the residual conductivity in the SC state, which is responsible partly for the reduced missing area in $\sigma_1(\omega)$.

A distinct difference from the μ SR results was recognized in the impurity-substituted YBCO,⁷ YBa₂Cu₄O₈ and many other disordered HTSC.⁵⁰ Disorder, impurity or defect, decreases the FIR SC condensate $(\lambda_L^{\text{FIR}})^{-2}$ at a much steeper rate than that expected from the linear $T_c - (\lambda_L^{\mu\text{SR}})^{-2}$ relationship observed by μ SR. For example, 0.4% Zn-substitution for optimally doped YBCO suppresses T_c from 93 K to 85 K and duplicates λ_L^{FIR} (~340 nm for $E \parallel a$), while $\lambda_L^{\mu\text{SR}}$ is expected to increase only 20% at most by the same Zn-substitution.⁵¹ The discrepancy between the FIR and μ SR data seems to be universal in the disordered or inhomogeneous cuprates, including the underdoped cuprates for which STM suggests an inherent spatial modulation of the SC order parameter over a length scales of a few nm. It may follow that the FIR measurement is more sensitive to microscopic inhomogeneity than μ SR, and that the ordinary relationship between ρ_s and λ_L ($\rho_s \sim \lambda_L^{-2}$) no longer holds for such microscopically inhomogeneous SC-states.

B. Low- ω residual conductivity at $T \ll T_c$

Finally we discuss the residual conductivity in the SC state. The Drude-like up-turn of conductivity towards $\omega=0$ is observed almost in all HTSC even at *T* well below T_c .⁹ In the present study, the low- ω Drude-like spectral weigh at $T \ll T_c$ is smaller for x=0.12 than for x=0.15, the Drude-like peak width being narrower in the former. This is consistent with the general trend that the spectral weight of the residual conductivity increases with doping.⁵²

The low- ω conductivity remaining at $T \ll T_c$ is an indication of the presence of the nonsuperconducting region. So far known are three types of inhomogeneous states in HTSC: (i) the presence of a non-SC region within the SC area, either metallic as speculated for overdoped cuprates by FIR (Ref. 53) and μ SR measurements,^{54,55} or pseudogapped as observed by STM for underdoped BSCCO;¹ (ii) alternating array of antiferromagnetic (AF) and SC stripes;⁵⁶ (iii) local suppression of the SC order around impurities such as Zn, as was directly observed by STM.⁵⁷

The impurity-induced inhomogeneity [case (iii)] has been investigated by many experimental techniques. In the optical spectra, Zn-substitution creates a huge residual conductivity, which dramatically depresses the missing area.⁷ A similar observation was reported for the Zn-doped YBa₂Cu₄O₈ (Ref. 50) and in the irradiated YBCO,⁵⁸ giving evidence for the sensitiveness of the FIR probe to microscopic inhomogeneity.

The case (ii), the spin and charge stripe order can be considered as an "ordered" inhomogeneous state, which has been detected by neutron scattering experiment as a static order in (La,Nd,Sr)₂CuO₄,⁵⁶ and probably as a dynamical form in LSCO,⁵⁹ where the Sr-content $x=0.12(\sim 1/8)$ is expected to be closer to the static order. In this case, the charge stripes on which SC order might develop are separated by the AF spin domains, and would form a periodic Josephson coupled array. A weak spatial modulation of the Josephson coupling strength between the stripes, due to e.g. stripe meand ering, is a possible source of the low- ω conductivity peak, as was observed in the *c*-axis optical response. $^{60-62}$ When the Josephson coupling strength is increased and strongly modulated spatially by decreasing an average spacing between stripes and/or by enhanced stripe fluctuation, the weight of residual conductivity would increase and be distributed over a fairly wide frequency range.

V. CONCLUSION

The in-plane polarized spectra of LSCO with x=0.12 and 0.15 were carefully measured. The peak at ~500 cm⁻¹ that was observed in the previous reports and/or in some of our samples was assigned to the *c*-axis phonon mode (A_{2u}) . Various possible sources for this admixture of *c*-axis spectral component were examined. Inaccurate angle of crystal cutting and/or multidomains in the TSFZ-crystals possibly introduces some amount of the dielectric response for $E \parallel c$. Measurement on *ab*-face with nonpolarized light or with *p*-polarization geometry also results in the *c*-axis component mixing. In any case, the problem originates from the large anisotropy in the electronic system of LSCO where only a small amount of *c*-axis component makes a serious effect on the in-plane spectrum, and therefore, this is the problem peculiar to a strongly anisotropic material like LSCO.

The obtained pure a/b-axis spectrum showed a clear superconducting response, the suppression of $\sigma_1(\omega)$ and $1/\omega$ -behavior of $\sigma_2(\omega)$. The estimation both from σ_1 and σ_2 give a consistent value of SC condensate, which indicates that the kinetic energy contribution is not appreciable. We find that the SC condensate is much smaller than that determined by μ SR. This discrepancy is possibly caused by microscopic inhomogeneity in the electronic state of superconducting CuO₂-planes, probably related to the stripe fluctuation in the case of LSCO. It is guessed that FIR-measurement is a probe sensitive to disorder which suppresses the SC order parameters over a length scale of nanometer. The inhomogeneous electronic state also seems to manifest in the spectrum as a residual Drude-like response at very low frequencies below 50 cm⁻¹ in the SC state.

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