I. INTRODUCTION

The mechanism of the normal-state charge transport remains to be one of the central issues in the studies of the high-$T_c$ cuprates, because it is intimately tied to the peculiarities of the strongly-correlated electron system in which the high-$T_c$ superconductivity is realized. The contrasting behavior between the in-plane resistivity $\rho_{ab}$ and the $c$-axis resistivity $\rho_c$ is arguably the most unusual property of the charge transport in the cuprates (Ref. 1); namely, there is an intriguing coexistence of a metallic behavior in $\rho_{ab}(T)$ ($d\rho_{ab}/dT > 0$) and an “insulating” (or semiconducting) behavior in $\rho_c(T)$ ($d\rho_c/dT < 0$) in many cuprates, and such an unusual nature of the anisotropic charge transport appears to offer a key to understand the charge transport mechanism. In fact, already a large amount of works have been concentrated on this issue. Probably the most important feature in the $c$-axis transport is that the magnitude of $\rho_c(T)$ is orders of magnitude larger than that expected from the band-structure estimates. This indicates that there is some “charge confinement” mechanism that renormalizes the $c$-axis hopping rate to a much smaller value. It has been argued that such “confinement” gives evidence for a non-Fermi-liquid nature of the cuprates, and the persistence of the contrasting behavior between $\rho_{ab}(T)$ and $\rho_c(T)$ down to very low temperatures does indeed suggest that some unconventional mechanism is responsible for the charge confinement, since any Fermi-liquid-based mechanism that provides an apparent confinement at finite temperatures should saturate at low enough temperature.

While it is likely that some unconventional mechanism (possibly a non-Fermi-liquid ground state) is fundamentally responsible for the confinement of charges into the CuO$_2$ planes (i.e. renormalization of the $c$-axis hopping rate), recent studies have shown 2-5 that a combination of the pseudogap and the $k$-dependence of the $c$-axis matrix element is largely responsible for the steeply insulating behavior of $\rho_c(T)$ observed, in particular, in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi-2212); namely, the angle-resolved photoemission spectroscopy (ARPES) measurements of Bi-2212 have demonstrated 3,4 that the pseudogap causes destruction of the Fermi surface starting from the $(0, \pm \pi)$ and $(\pm \pi, 0)$ points, and the $c$-axis matrix-element effect 3,4 tends to amplify the contribution of the electrons on these gapped portions of the Fermi surface to the $c$-axis transport. As a result, the $c$-axis conductivity is quickly diminished upon opening of the pseudogap; the steep upturn in $\rho_c(T)$ observed in YBa$_2$Cu$_3$O$_{6+y}$ (YBCO), Bi-2212, and Bi$_2$Sr$_2$CuO$_{6+\delta}$ (Bi-2201) below the pseudogap temperature is considered to be due to this mechanism. On the other hand, it has been shown by the ARPES measurements that the Fermi surface of the La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) system appears to be quite different from that of Bi-2212 (the stripe fluctuations cause the Fermi surface to be one-dimensional (1D) like YBCO), and it is not likely that the pseudogap shows a similar development on the Fermi surface of the LSCO; therefore it would be illuminating to examine the $c$-axis transport of LSCO, particularly in the underdoped region, and compare it with that of other cuprate systems.

In the past, the $c$-axis transport in LSCO has already been rather well studied through various techniques such as resistivity, 6-8 magnetoresistance, 9-11 microwaves, 12 and optics. 13,14 This is partly because large single crystals can be grown with the traveling-solvent floating-zone (TSFZ) technique for LSCO and large single crystals can be grown with the traveling-solvent floating-zone (TSFZ) technique for LSCO and thus it is easy to obtain samples that are long enough along the $c$-axis to allow reliable $c$-axis transport measurements. It has been demonstrated 15,16 that $\rho_c(T)$ in slightly underdoped samples ($0.10 < x < 0.16$) shows only a weak temperature dependence at moderate...
temperatures (100 – 300 K), particularly after correcting for the thermal expansion,
which is in contrast to the behaviors of \( \rho_c(T) \) of Bi-2212 or Bi-2201,
where the pseudogap causes \( \rho_c(T) \) to become steeply insulating.
The magnetoresistance study by Hussey et al. has shown that the \( c \)-axis transport of LSCO is
incoherent and \( \rho_c \) is governed in a large part by the
in-plane scattering rate. The low-temperature normal-
state behavior of \( \rho_c(T) \) in LSCO has been found to track
the behavior of \( \rho_{ab} \), namely, both \( \rho_{ab}(T) \) and \( \rho_c(T) \) for
\( 0.08 \leq x \leq 0.15 \) show a peculiar localizing behavior with
the same temperature dependence of \( \log(1/T) \)
(Refs. 21,23); this seems to be in good accord with
the magnetoresistance result of Hussey et al. which
suggests that the strong in-plane scattering can cause the carriers to localize in both the \( ab \) and \( c \) directions.

However, all the \( c \)-axis transport studies that used the
TSFZ-grown single crystals have been limited to the region
with \( x \geq 0.06 \), and thus it is important to expand the region of \( x \) to smaller values, down to the lightly-
doped region, to establish a complete picture of the \( c \)-axis transport in LSCO. Since we have recently succeeded in
growing a series of high-quality single crystals of LSCO from \( x = 0.01 \) to 0.1 and have measured their in-plane transport properties \( \rho_{ab} \) it is natural for us to study the
\( c \)-axis transport using those high-quality single crystals.
There were studies of the transport properties of flux-
grown \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) single crystals in the antiferromagnetic regime, but the data are inconsistent with the
present study (for \( \rho_c \)) and with Ref. 29 (for \( \rho_{ab} \)). Studies of the \( c \)-axis transport in the lightly-doped LSCO
would be particularly useful for clarifying the mechanism of the \( c \)-axis transport, in view of the unusually metallic behavior of the in-plane mobility which suggests that the holes are likely to form a self-organized network of hole-rich regions where a band-like metallic transport occurs.(Such a phase has been theoretically predicted to be realized in correlated electron systems and was named “electronic liquid crystal”.) Also, recent ARPES measurements have provided information on the Fermi surfaces of lightly-doped LSCO, which is useful in interpreting the transport data.

In this paper, we detail our technique to grow high-
quality LSCO single crystals and present the \( c \)-axis transport data taken on the crystals with \( x = 0.01 \) – 0.10.
The corresponding \( \rho_{ab}(T) \) data for the same compositions are also presented for the analysis of the resistivity anisotropy. It is found that the resistivity anisotropy ratio \( \rho_c/\rho_{ab} \) at moderate temperatures becomes almost independent of \( x \) in the lightly-doped region; this is contrary to the common belief that the resistivity anisotropy should increase with decreasing doping and thus is a surprising result. We discuss that this striking \( x \)-independence of \( \rho_c/\rho_{ab} \) is consistent with the idea that holes are segregated into a network of hole-rich regions and that the \( c \)-axis transport is essentially an incoherent hopping between these self-organized networks, in which the strength of the “confinement” does not change with \( x \).

II. EXPERIMENTAL DETAILS

A. Crystal Growth and Preparation

The series of \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) single crystals (0.01 \( \leq x \leq 0.10 \)) are grown by the TSFZ technique. Raw powders of \( \text{La}_2\text{O}_3, \text{SrCO}_3, \) and \( \text{CuO} \) (with purities of 99.9% or higher) are dried at 600°C for 12 hours prior to weighing, and then are weighed and mixed with a composition in which \( \text{CuO} \) is 2 – 3% richer than the nominal one; the additional \( \text{CuO} \) works as a binder of the polycrystalline feed-rod and helps avoiding the occurrence of cracks in the rod, as well as avoiding penetration of the molten liquid into it, during the TSFZ operation. Also, since the vapor pressure of \( \text{CuO} \) during the growth is relatively high and thus the resulting crystals should have smaller \( \text{Cu} \) content compared to the starting material, the excess \( \text{CuO} \) in the feed-rod compensates the \( \text{Cu} \) loss and is necessary to obtain stoichiometric single crystals as the final product.

The mixture of raw powders is well ground and calcined at 750°C for 12 hours, then at 920°C for 12 hours for 4 times in alumina crucibles with regrinding between each calcination. After the last calcination and regrinding, the powders are isostatically pressed into a rod shape with typical dimensions of 7 mm \( \times \) 150 mm, and finally sintered at 1200°C for 15 hours in air to form the feed-rod. Calcination and regrinding for more than 4 times before the final sintering make the polycrystalline materials highly homogeneous, and this homogeneity is important for the sintered feed-rod to become dense and hard, which ensures the molten zone to be well stabilized during the TSFZ operation.

The solvent material, which is used to lower the temperature of the molten zone, is prepared to have the cation ratio of \( \text{La} : \text{Sr} : \text{Cu} = (2 – x) : x : 3 \) for each \( x \). We use 0.5 g of the solvent for the TSFZ growth. The TSFZ operation is carried out using an infrared image furnace (NEC Machinery SC K-15HD) with two halogen lamps and double ellipsoidal mirrors in the atmosphere of flowing dried air. To cut off reflections of the light from high angles, a quartz tube (which encloses the moving rods) is partly covered with aluminum foils to make a narrow window; this sharpens the temperature profile and helps stabilizing the molten zone. Use of a seed crystal helps the whole rod to become a single crystal. The growth rate is kept constant at 1.0 mm/h (or less). A photograph of our TSFZ-grown crystal with \( x = 0.04 \) is shown in Fig. 1(a) as an example, together with its X-ray rocking curve [Fig 1(b)]. The full-width-at-half-maximum (FWHM) of the (006) rocking curve shown in Fig. 1(b) is only 0.1°, which is the smallest reported value for LSCO single crystals. (The X-ray beam is always wide enough to cover the whole specimen when the rocking curve is taken.)
The samples with $x \leq 0.05$ are carefully annealed in pure Ar gas at 400°C for 20 hours to remove excess oxygen. It should be noted that in the lightly-doped region the transport properties are strongly affected by the excess oxygen; even a slight amount of excess oxygen can easily cause phase-separated oxygen-rich regions which show superconductivity. (Many of the early data of lightly-doped LSCO, like those of Ref. 34, are actually contaminated by the superconductivity caused by the excess oxygen.) We have examined various annealing conditions and found that annealing at higher temperature and/or for longer time in pure Ar does not change the resistivity any more; therefore, we have concluded that the above annealing is sufficient to get rid of the excess oxygen while minimizing the possible decomposition. On the other hand, the samples with $x = 0.08$ and 0.10 are annealed at 800°C for 40 hours in air, followed by rapid quenching to room temperature, to remove oxygen defects that cause additional electron scattering in moderately-doped and overdoped samples.

After the growth is finished, the whole chunk of crystal is annealed at 850°C for 20 hours in air to remove thermal stresses formed during the FZ growth. The crystallographic axes are precisely determined by the X-ray back-reflection Laue method, which also reveals that the typical growth direction is [110] (in the notation of high-temperature tetragonal phase). The samples for the present measurements are shaped into thin platelets with typical dimensions of $2.0 \times 0.5 \times 0.2$ mm$^3$, with the wide faces exactly parallel to the $ab$ ($ac$) planes within an error of 1° for the in-plane (out-of-plane) measurements.

The in-plane and out-of-plane resistivities ($\rho_{ab}$ and $\rho_c$) are measured using a standard ac four-probe method. Note that, since we can obtain samples that are long along the $c$-axis, we do not need to use a complicated technique, such as the Montgomery method, for the $\rho_c$ measurement in the case of LSCO. The contact pads are hand-drawn on polished sample surfaces with gold paint, followed by a heat treatment at 400°C for 30 minutes in air, which makes the gold particles to well adhere to the crystal surface [the high-temperature annealing to remove excess oxygen (or oxygen defects) are done after this process]. For the current contacts, the whole area of two opposing side faces is painted with gold to ensure uniform current flow through the sample. After the annealing, thin gold wires are attached to the contact pads using silver epoxy, which electrically and mechanically binds the wire to the sample; the contact resistance with this technique is less than 1 Ω.

The photographs in Figs. 1(c) and 1(d) show the geometry of the contacts and how the leads are attached.

The uncertainty in the absolute values of $\rho_{ab}$ and $\rho_c$ is minimized by using relatively long samples (at least 2 mm long) and by painting narrow contact pads with a width of 50–80 μm; total errors in the absolute values are less than 10%. We note that, although the measurements employ a simple four-probe method, each sample has four voltage contacts (two on each side face) and the resistivity is measured using both pairs [see Fig. 1(d)]. Sometimes the resistivity values from the two voltage pairs do not agree, which is an indication of some inhomogeneity (or microcrack) in the sample; we reject the data when such a discrepancy is observed, and all the data shown here are measured on samples for which the two data sets match within 5%. We also confirm that the resistivity data measured on different pieces of crystals from the same batch are reproducible within 10% as long as the samples are free from inhomogeneity.

### III. RESULTS

Figure 2 shows the temperature dependences of $\rho_{ab}$ of our crystals with the vertical axis in the logarithmic scale; this figure is essentially a partial reproduction of the data from Ref. 29, except for the $x=0.10$ data that are new. The absolute value of $\rho_{ab}$ is among the smallest ever reported for each $x$. This fact, together with the very sharp X-ray rocking curve, indicates that the single crystals studied here are of very high quality. As was emphasized in Ref. 29, the temperature dependence of $\rho_{ab}$ is metallic ($d\rho_{ab}/dT > 0$) at moderate temperatures even in the samples where $\rho_{ab}$ exceeds 2 mΩcm, which corresponds to $k_Fl$ values of less than 1, and thus the Mott-Ioffe-Regel limit for metallic transport is strongly violated; we have argued that this behavior, in combination with the mobility that is only weakly
doping-dependent, is best understood to result from a self-organized network of hole-rich regions that constitute the path for the charge transport.

Figure 3 shows the temperature dependence of $\rho_c$ for the same doping range. In the $\rho_c(T)$ profile of LSCO, a clear kink is usually observed at the structural phase transition temperature from the high-temperature tetragonal (HTT) phase to the low-temperature orthorhombic (LTO) phase for all the compositions of the present study, however, the structural phase transition occurs at temperatures higher than 300 K (Ref. 35), except for the $x=0.10$ sample which show the LTO transition at 293 K. We note that the data for $x=0.01$ show a kink at 240 K, which corresponds to the Néel temperature $T_N$ and not to the LTO transition temperature. (The kink in the $\rho_c(T)$ curves at $T_N$ has been reported for antiferromagnetic YBCO, but not for LSCO before.) It is useful to note that, although $\rho_c$ increases with decreasing temperature in all the samples, the temperature dependence of $\rho_c$ is rather weak at moderate temperatures (for example, $\rho_c$ of the $x=0.02$ sample shows only a factor of 2 increase from 300 K to 50 K), and a negative curvature ($d^2\rho_c/dT^2 < 0$) is observed in the $\rho_c(T)$ profile for $x = 0.02$ and 0.03 at temperatures above 100 K (which can be seen in Fig. 4). This negative curvature is in sharp contrast to the steeply positive curvature of $\rho_c(T)$ of Bi-2212 (Ref. 13) or Bi-2201 (Ref. 17) below the pseudogap temperature. At lower temperatures below 50 K, the $\rho_c(T)$ curves of all the samples show strongly diverging behavior, which is in clear correspondence with the localization behavior in $\rho_{ab}(T)$ at low temperatures (except for the $x=0.08$ and 0.10 samples whose $\rho_{ab}(T)$ curves show an insulating behavior only when the superconductivity is suppressed by a high magnetic field). An intriguing aspect of the $\rho_c(T)$ data is that their profile does not change much with $x$ (namely, the $\rho_c(T)$ data are almost parallel-shifted for different $x$) at moderate temperatures, as is the case with the $\rho_{ab}(T)$ data. To examine the doping dependence of the $\rho_c(T)$ data in detail, we plot the temperature dependences of $n_h e \rho_c$, which corresponds to the inverse mobility $\mu_{c}^{-1}$ of the doped holes along the $c$-axis, where $e$ is the electronic charge and $n_h$ is the nominal hole concentration given by $2x/V$ (unit cell volume $V$ is $3.8 \times 3.8 \times 13.2$ Å$^3$), as we did for $\rho_{ab}(T)$ in Ref. 29. As is shown in Fig. 4, the absolute magnitude of $n_h e \rho_c$ at 300 K changes only by $\sim 40\%$ in the non-superconducting regime (from $x = 0.01$ to 0.05), although it starts to change quickly in the superconducting regime (above $x=0.05$). The weak $x$-dependence of $n_h e \rho_c$ at 300 K in the non-superconducting regime is rather similar to the behavior of the inverse in-plane mobility $\mu_{ab}^{-1}$ that also changes only by $\sim 40\%$ from $x = 0.01$ to 0.05 (Ref. 29).

The most surprising feature we find in this work is that $\rho_c/\rho_{ab}$ at moderate temperatures is almost completely independent of doping in the non-superconducting regime.
(0.01 ≤ x ≤ 0.05), as illustrated in Fig. 5. When x exceeds 0.05 and enters into the superconducting regime, ρc/ρab starts to become smaller with increasing x. (The implication of this result is discussed in detail in the next section.) Also, all the samples in Fig. 5 show strongly temperature-dependent ρc/ρab, and in the non-superconducting samples (x ≤ 0.05) there is a peak that moves systematically to higher temperature with decreasing x. When one compares Fig. 5 to Fig. 2, it becomes clear that the peak temperature of the anisotropy corresponds to the onset of the insulating behavior in ρab(T); namely, the decrease of ρc/ρab with decreasing temperature is determined by the rapid increase in ρab. The occurrence of the peak in ρc/ρab means that the strong localization of the carriers causes ρc to diverge more rapidly than ρab, and as a result the system becomes less two-dimensional in the strongly localized state; this can be rephrased that the strong localization causes the system to become a “three-dimensional” insulator where the carriers cannot move to any direction.

It is worth noting that it is not the establishment of the long-range antiferromagnetic order but the strong localization effect that cause ρc/ρab to decrease; in this regard, our result disagrees with the interpretation of Kitajima et al. for the ρc/ρab behavior of insulating Bi2Sr2ErCu2O8. Our sample with x=0.01 clearly shows that ρc/ρab keeps increasing with decreasing temperature (which means that the charge confinement becomes increasingly effective) even below the Néel temperature until the carriers start to localize.

IV. DISCUSSIONS

As is seen in Fig. 3, the temperature dependence of ρc at moderate temperatures (100 – 300 K) is weak for x ≥ 0.02, which is in contrast to the ρc(T) behaviors of Bi-2212 or Bi-2201 where ρc(T) shows a steep upturn below the pseudogap temperature [a steep upturn in ρc(T) of LSCO is only observed at low temperature (below ∼50 K), which is most likely to be associated with the strong localization]. Since the c-axis transport in any cuprate should necessarily reflect the c-axis matrix element that tends to amplify the development of the pseudogap with a d_{x^2−y^2} symmetry, the ρc(T) behavior of underdoped LSCO means either that the pseudogap is already fully developed at 300 K, or that the pseudogap has a symmetry different from d_{x^2−y^2}. (The photoemission experiment has observed a pseudogap in LSCO only in the angle-integrated mode, and the temperature evolution was not studied). Note also that the published ARPES results to date have reported that the Fermi surface in the underdoped LSCO is observed only near the (±π, 0), (0, ±π) points (1D-like Fermi surface), which is incompatible with the concept of the pseudogap with a d_{x^2−y^2} symmetry; this fact might also be related to the weak temperature dependence of ρc at moderate temperatures. (It should be noted, however, that very recent ARPES measurements of the lightly-doped LSCO have succeeded in observing a developing band at the nodal points so
the picture of the Fermi surface of LSCO is not finalized yet.) In any case, we expect that future ARPES measurements will help understanding the behavior of $\rho_c(T)$ in LSCO.

The most illuminating observation is that the resistivity anisotropy $\rho_c/\rho_{ab}$ near room temperature is almost independent of $x$ for 0.01 $\leq$ $x$ $\leq$ 0.05 (Fig. 5). In this $x$-independent regime, the magnitude of $\rho_c/\rho_{ab}$ is $\sim$1000 at 300 K and increases with decreasing temperature, which indicates that the same charge confinement mechanism is at work down to $x$=0.01; interestingly, this range of $x$ (0.01 – 0.05) corresponds to the region where the neutron experiments have found diagonal spin stripes. As was already mentioned in the previous section, the metallic in-plane transport that violates the Mott-Ioffe-Regel limit suggests that the transport is occurring through a self-organized network of hole-rich regions that constitute the path for the charge transport. The striking $x$-independence of $\rho_c/\rho_{ab}$ is naturally understood in this picture of transport through a self-organized network, because in such a case the transport anisotropy is determined by the local electronic nature of the hole-rich segment that is presumably unchanged with $x$ (only the average distance between the hole-rich paths changes with $x$). Therefore, the behavior of $\rho_c/\rho_{ab}$ in the lightly-doped region is consistent with the self-organized-network picture and gives strong support to it.

It should be noted that the $c$-axis mobility, as well as the $\rho_{c}/\rho_{ab}$ ratio, quickly changes when $x$ enters into the superconducting regime. As can be seen in Fig. 4, $n_x e \rho_{c}$ at 300 K changes by a factor of 3 from $x$=0.05 to 0.10, which indicates that the $c$-axis hopping becomes easier as $x$ is increased above 0.05. Correspondingly, the anisotropy ratio at moderate temperatures starts to show a decrease above $x$=0.05, which means that the charges are increasingly less confined in this doping range. This sharp change in the $c$-axis transport properties across $x$=0.05 is in contrast to the smooth change of the inplain transport properties, where the mobility at 300 K changes very smoothly as $\sim \sqrt{x}$ (Ref. 29), neglecting the superconductor-to-insulator transition that occurs at low temperature. As we discussed in Ref. 29, the fact that the in-plane mobility changes slowly and smoothly and is governed by a simple function of $x$ from $x$ = 0.01 to 0.17 strongly suggests that the mechanism of the in-plane charge transport is essentially unchanged from the antiferromagnetic regime to optimum doping; since the transport in the antiferromagnetic regime is likely to be governed by a self-organized network (which can be interpreted to be a generalized version of the “stripes”),

the charge transport in the whole phase diagram up to optimum doping appears to be governed by such an electron self-organization, which also gives a natural picture for the charge confinement. The difference in the $x$ dependence between $\mu_{c}$ and $\mu_{ab}$ probably indicates that the $c$-axis coupling (or correlation) between the two-dimensional networks is more susceptible to the change in $x$ than the properties of the network itself. It is intriguing to note that the emergence and increase of $T_c$ appear to be related to an increase in the $c$-axis coupling between the self-organized network.

V. SUMMARY

The $ab$- and $c$-axis resistivities are measured in the lightly- to moderately-doped LSCO single crystals and the resistivity anisotropy is analyzed to sort out the transport mechanism of this prototype cuprate. At moderate temperatures, the temperature dependence of $\rho_{c}$ in the underdoped LSCO is much weaker than that of underdoped Bi-2212 or Bi-2201; we discuss that this difference in the $\rho_{c}(T)$ behavior is related to the differences in both the Fermi-surface topology and the way how the pseudogap develops on the Fermi surface. Moreover, we found that $\rho_{c}/\rho_{ab}$ near room temperature is almost completely independent of doping for 0.01 $\leq$ $x$ $\leq$ 0.05. This result supports the picture that the holes form a self-organized network of hole-rich path, which was originally suggested from the unusually metallic in-plane transport of the holes in the lightly-doped region; in this picture, it is understood that the band-like in-plane transport takes place through the self-organized network, while the $c$-axis transport is essentially an incoherent hopping between the networks, in which the strength of the confinement does not change with $x$ in the lightly-doped region. Our data for $x > 0.05$ suggest that the weakening of the $c$-axis charge confinement is related to the emergence of the superconductivity, while the in-plane transport mechanism appears to be unchanged from $x$=0.01 to optimum doping.

Acknowledgments

We thank A. Fujimori and S. A. Kivelson for helpful discussions, and T. Sasagawa and K. Kishio for valuable suggestions for the crystal growth. X.F.S. acknowledges support from JISTEC.
