Zn-doping effects on the electrical resistivity of $La_{1.85}Sr_{0.15}CuO_4$ under a magnetic field

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We measured the electrical resistivity of $La_{1.85}Sr_{0.15}Cu_{1-y}Zn_yO_4$ ($y=0,0.004,0.008,0.017$) single crystals to study the magnetic-field dependence of the superconducting transitions. For $y=0\sim0.008$, as the magnetic field along the c axis is increased, the superconducting transition significantly broadens while the onset T_c remains unchanged in both the in-plane resistivity ρ_{ab} and the out-of-plane resistivity ρ_c. This corresponds to the so-called *broadening behavior*. For $y=0.017$, the onset T_c is apparently reduced with increasing the magnetic field, which we define as a *parallel shift*. This indicates that the in-plane superconducting coherence length drastically increases at $y=0.017$. We discuss these Zn-doping effects based on an inhomogeneous picture.

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I. INTRODUCTION

Early studies have shown that the electrical resistivity under a magnetic field exhibits a characteristic behavior in the high-T_c superconductors: in the underdoped regime, the superconducting transition width ΔT_c broadens as the magnetic field along the c axis increases, while the onset T_c remains almost unchanged.[1–4] This is called a *broadening behavior*, which is now mainly ascribed to "superconducting fluctuations" and "vortex motion." The superconducting fluctuations indicate that the superconducting order parameter fluctuates near T_c. The fluctuations become large in the high-T_c superconductors because of their short superconducting coherence lengths ξ and low dimensionalities.[5] The vortex motion means that the Lorentz force drives vortices under a magnetic field so that the electric resistivity is induced.

Recently, it was reported that such a broadening behavior is eliminated for a certain kind of material; in $La_{2-x}(Nd,Sr,Ba)_xCuO_4$ around $x=1/8$, the onset T_c is significantly decreased by a magnetic field, while ΔT_c remains constant.[6–8] We refer to this behavior as a *parallel shift*. The materials, which exhibit the parallel shift, show a specific phenomenon; neutron scattering studies have revealed incommensurate (IC) orders due to spin correlations and most likely due to charge correlations as well. Such a parallel shift might be characteristic to a case where the IC static correlations appear. Although both the broadening behavior and the IC spin fluctuations are typical phenomena in the high-T_c superconductors, the parallel shift has been observed only for the hole doping $x\sim1/8$. Therefore, a qualitatively different system is required to clarify whether the appearance of the IC static correlations and the parallel shift is intrinsically related or accidentally coincident.

In this paper, we report on the electrical resistivity measurements around T_c under a magnetic field for $La_{1.85}Sr_{0.15}Cu_{1-y}Zn_yO_4$ ($y=0,0.004,0.008,0.017$) single crystals. It is known that impurity-free $La_{1.85}Sr_{0.15}CuO_4$ shows a "spin gap" in the antiferromagnetic (AF) IC spin excitations at low temperatures and that no static magnetic correlation have been observed for $y=0$.[9–11] Recent studies have shown that the spin gap is gradually filled and that the IC static magnetic correlations appear by substituting Zn for Cu.[12–14] The purpose of the present work is to investigate whether the resistivity curve near T_c changes with the appearance of static magnetic correlations. We systematically measured both $\rho_{ab}(H\perp J\parallel ab)$ and $\rho_c(H\parallel J\parallel c)$ for various Zn concentrations under a magnetic field along the c axis.

II. EXPERIMENTAL

Single crystals of $La_{1.85}Sr_{0.15}Cu_{1-y}Zn_yO_4$ ($y=0,0.004,0.008,0.017$) were grown using the traveling-solvent-floating-zone method. The concentrations of La, Sr, Cu, and Zn ions were precisely estimated by the inductively coupled plasma analysis. In addition, we measured the superconducting shielding signals with a superconducting quantum interference device magnetometer to determine bulk T_c. Each sample except for $y=0$ was cut from the same crystal rod as used in previous neutron scattering measurements,[12–14] and the details of crystal growth and characterization are given elsewhere.[14] As for $y=0$, the estimated Sr concentration is $x=0.142(2)$ and the shielding signal shows a sharp transition at T_c (midpoint) of 37.4 K. The in-plane (ρ_{ab}) and out-of-plane (ρ_c) resistivities were measured by a standard four-probe dc method using a rotating sample holder to precisely align the samples against the magnetic field. We prepared long rectangular crystals, which sizes are typically $0.7\times0.7\times5$ mm^3 for ρ_{ab} and $1\times1\times3$ mm^3 for ρ_c. After a silver paste was painted, the samples were heated under oxygen gas flow at 350–500 °C for 2 h to obtain a low contact resistance. The whole areas of both the ends were painted with a silver paste to assure a uniform current flow through the sample and gold wires were used as lead. After these processes, the contact resistance became less than 1 Ω. The samples were first cooled to the lowest temperature under magnetic field along the c axis up to 10 T. After the sample temperature was stabilized at each measuring point in a heating process, the resistivity measurements were repeated ten times and the data were averaged. Electric currents used for measurements were 10 mA for ρ_{ab} and 1 mA for ρ_c to ensure the accuracy for voltage reading. The Joule heating to the sample at low temperatures is esti-

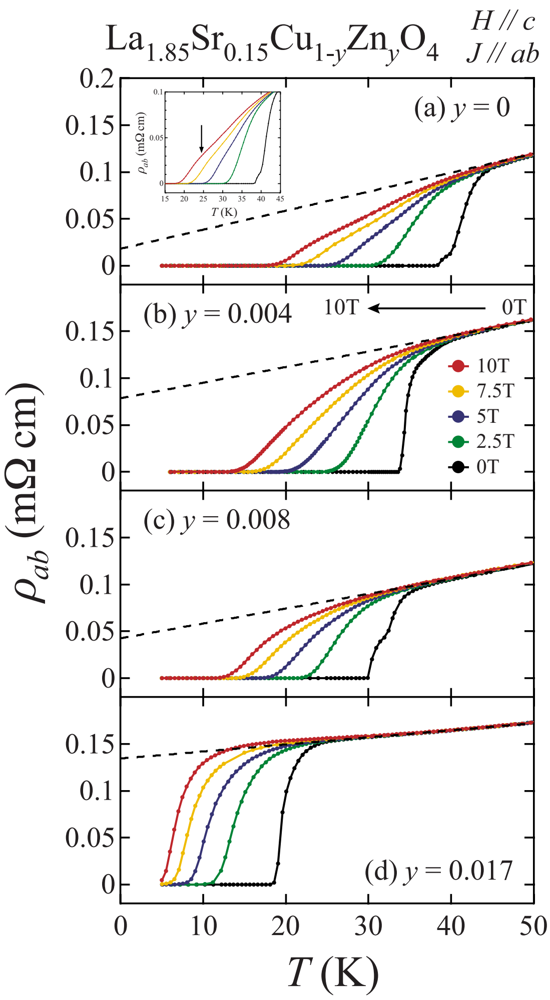

FIG. 1. (Color online) Temperature dependence of ρ_{ab} for $La_{1.85}Sr_{0.15}Cu_{1-y}Zn_yO_4$ with (a) $y=0$, (b) $y=0.004$, (c) $y=0.008$, and (d) $y=0.017$. Magnetic fields were applied along the c axis from 0 to 10 T. The resistivity in the normal state ρ_{normal} is estimated by the extrapolation from the linear T-dependent region just above T_c at $H=0$ T. Dashed lines represent the estimated resistivity in the normal state. The inset shows the enlarged view of ρ_{ab} for $y=0$. The arrow in the inset denotes the temperature at which a kink in the ρ-T curve occurs.

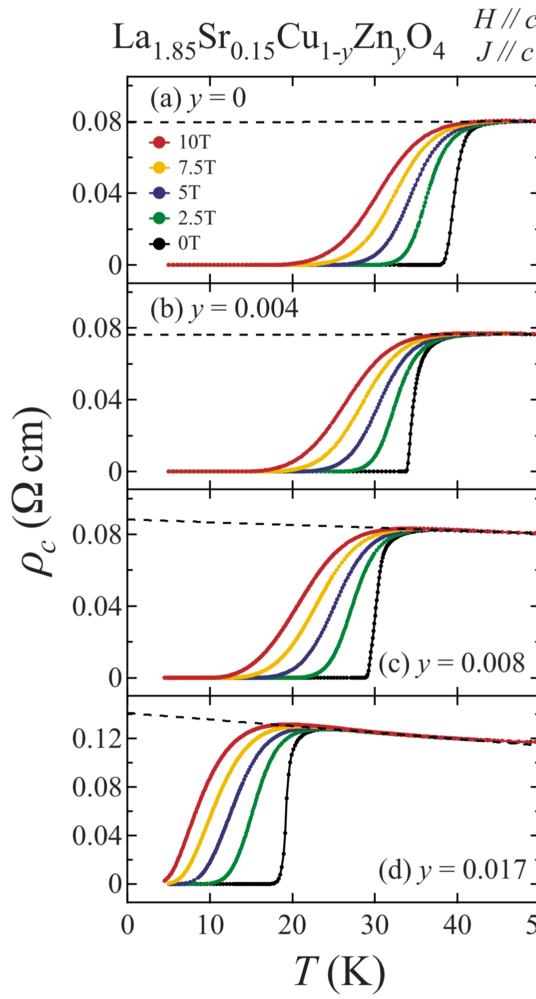

FIG. 2. (Color online) Temperature dependence of ρ_c for $La_{1.85}Sr_{0.15}Cu_{1-y}Zn_yO_4$ with (a) $y=0$, (b) $y=0.004$, (c) $y=0.008$, and (d) $y=0.017$. Magnetic fields were applied along the c axis from 0 to 10 T. The resistivity in the normal state ρ_{normal} is estimated by the extrapolation from the linear T-dependent region just above T_c at $H=0$ T. Dashed lines represent the estimated resistivity in the normal state.

mated at 1 nW (10^{-6} W) or less, which increases the sample temperature at a rate of 0.6 K/s at most without any cooling. This should be easily compensated with the cooling power of the cryostat. The angle between a magnetic field and a current was confirmed by measuring the angle dependence of the resistivity.

III. RESULTS

Figures 1 and 2 show the temperature dependence of the in-plane resistivity ρ_{ab} and the out-of-plane resistivity ρ_c under magnetic fields along the c axis. The ρ_{ab} for $y=0$ and 0.008 in zero field show a two-step behavior, which probably comes from inhomogeneities of Sr or O in the sample. Since the transition at lower temperature approximately corresponds to the onset T_c in the magnetization, it is considered that the transition at higher temperature occurs due to accidental connections of current paths of superconductivity and that the transition at lower temperature shows the bulk superconductivity. As for $y=0$, the resistivity exhibits a typical broadening behavior as previously reported,[2–4] and the behavior in ρ_{ab} is marked compared with that in ρ_c. The broadening of ρ_{ab} is gradually suppressed by increasing Zn and ρ_{ab} undergoes a parallel shift for $y=0.017$. In addition, we observed a kink in the resistivity curve of ρ_{ab} for $y=0$ [the inset in Fig. 1(a)], and the kink gradually disappears with increasing Zn. However, no kink was observed in ρ_c for all the samples.

In Fig. 3, we show magnetic-field dependences of two temperatures: one is the temperature at which the resistivity decreases to 95% of the resistivity in the normal state $\rho_{\mathrm{normal}}(r=\rho/\rho_{\mathrm{normal}}=0.95)$, $T_c(r=0.95)$, which is defined as the onset T_c in this paper. The other is the temperature at $r=0.05$, $T_c(r=0.05)$, which corresponds to the end of transition. The difference between these two temperatures corre-

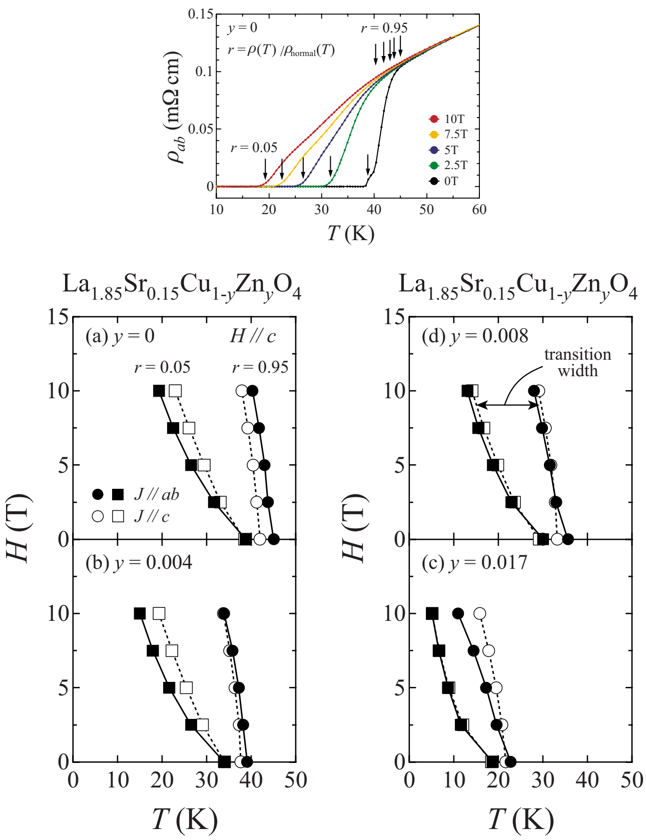

FIG. 3. (Color online) Magnetic-field dependences of temperatures at which the resistivity decreases to 95% (r=0.95) and 5% (r=0.05) of the resistivity in the normal state for $La_{1.85}Sr_{0.15}Cu_{1-y}Zn_yO_4$ with (a) y=0, (b) y=0.004, (c) y=0.008, and (d) y=0.017. Top figure shows the temperature at r=0.95 and 0.05 for the in-plane resistivity for y=0.

sponds to the transition width ΔT_c. Here we mention about an uncertainty of the estimated T_c. ρ_{normal} slightly changes with field, namely, magnetoresistance known as the Aslamazov-Larkin term and the Maki-Thompson term and we checked how it affects the estimation of T_c. We fitted the data taken at both H=0 and 7.5 T (or 10 T) with a linear function to determine ρ_{normal} and compared the results. In fact, $T_c(r=0.95)$ is somehow sensitive to the determination of ρ_{normal}, but its uncertainty was estimated to be less than 0.8 K, and the fitting error is much smaller than this uncertainty.

Obviously, we can see that the transition width at a high magnetic field becomes small with doping Zn, which would indicate the broadening behavior changes into the parallel shift. However, the transition width may not be a good indicator to distinguish the parallel shift from the broadening behavior. Because $T_c(r=0.05)$ must be positive value and tends to stagnate at higher magnetic fields as the transition temperature is low. Therefore, the transition width is seemingly reduced for the sample with low T_c. For the reason, we consider that the field dependence of onset T_c, $T_c(r=0.95,H)$, is better indicator than the transition width. For ρ_{ab} of y=0–0.008, $T_c(r=0.95)$ shows a weak magnetic-field dependence, which suggests the broadening. In contrast, $T_c(r=0.95)$ for y=0.017 apparently shifts to lower temperatures with increasing the magnetic field, indicating the parallel shift. The parallel shift behavior is marked in ρ_{ab} compared with that in ρ_c. Furthermore, we note that $T_c(r=0.05)$ of ρ_{ab} does not coincide with that of ρ_c for y=0. Upon doping Zn, these two values become closer and finally coincide for y=0.017. The resistivity near $T_c(r=0.95)$ can be described by superconducting fluctuations, while the resistivity near $T_c(r=0.05)$ can be affected by vortex motion. We, however, cannot simply distinguish between the effect by superconducting fluctuations and that by vortex motion. We discuss the Zn-doping dependence of $T_c(r=0.05)$ and $T_c(r=0.95)$ in the Sec. IV.

IV. DISCUSSIONS

First, we discuss the magnetic-field dependence of $T_c(r=0.05)$ in ρ_{ab} and ρ_c. As seen in Fig. 3, at higher fields, $T_c(r=0.05)$ in ρ_{ab} does not coincide with that in ρ_c for y=0. However, the difference becomes small upon doping Zn. Here we note that the gradient, dH/dT, becomes small as Zn increases in ρ_{ab} but shows no Zn-doping dependence in ρ_c. It appears that there is an additional component in ρ_{ab} and that the component is suppressed by Zn. Since the vortex motion originates from the Lorentz force, the resistivity contains the contribution of vortex motion only when the magnetic field is applied perpendicular to the current pass ($H \perp J$). In our measurement, ρ_{ab} is contributed from the vortex motion because $J \| ab$ is perpendicular to $H \| c$. Furthermore, in the inset of Fig. 1(a), we see kinks in the ρ-T curve at high magnetic fields. The previous study reported that the kink is attributed to a melting transition of the vortex lattice.[15] Moreover, a recent study for the $YBa_2Cu_3O_{7-\delta}$ shows that a vortex lattice phase disappears with doping Zn,[16] indicating the pinning of vortices by Zn. We speculate that the observed kink for y=0 corresponds to a vortex liquid–vortex glass (or short-range vortex lattice) transition and that the vortex glass state is stabilized upon doping Zn.

Next, we observed the parallel shift behavior in ρ_{ab} for y=0.017 unlike y=0–0.008, indicating that the superconducting fluctuations are small for y=0.017. The parallel shift was observed in particular materials, such as $La_{2-x}(Nd,Sr,Ba)_xCuO_4$ around x=1/8, which are expected to have static IC magnetic correlations or both charge and magnetic IC correlations.[6–8] In our samples, the elastic IC magnetic signals were observed for y=0.017, while no signals appear for y=0–0.008.[12–14] Thus, we conclude that the parallel shift is intrinsically related to the static IC correlation in the Zn-doped $La_{1.85}Sr_{0.15}CuO_4$ and that these phenomena are observed not only in the high-T_c materials around the 1/8 hole concentration[6–8] but also in impurity-doped systems. It indicates that the transport properties in the CuO_2 plane have some relationship to the magnetic one.

Let us discuss the Zn-doping dependence of superconducting coherence length ξ. According to the superconducting fluctuation theory with taking account of the $|\psi|^4$ term in the Ginzburg-Landau equation, the resistivity can be induced below the mean-field critical temperature $T_c(H)$.[5] It was also reported that the onset temperature of diamagnetism was quite different from the R=0 temperature, which seems more

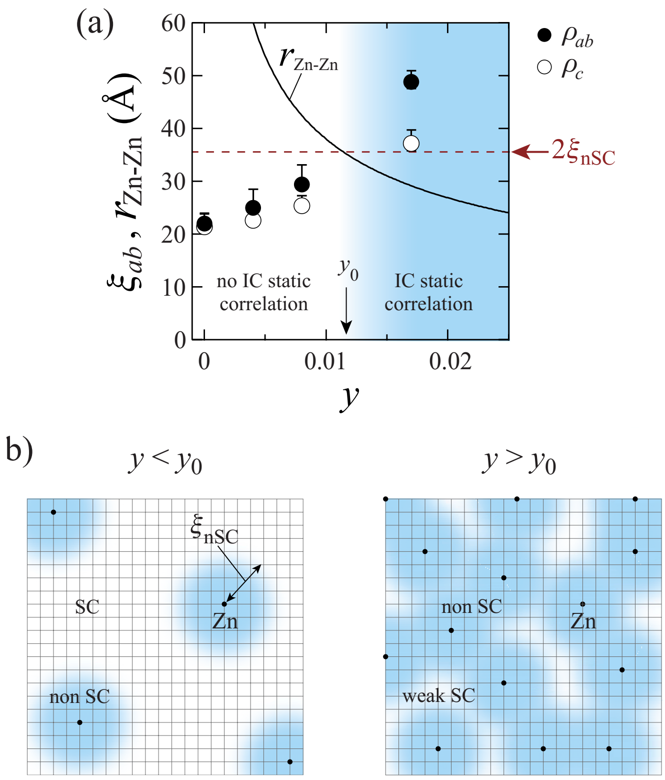

FIG. 4. (Color online) (a) In-plane superconducting coherence lengths estimated from $T_c(r=0.95)$ of ρ_{ab} (solid circles) and ρ_c (open circles) for $y=0$, 0.004, 0.008, and 0.017. The coherence lengths from $T_c(r=0.98)$ and $T_c(r=0.90)$ are represented as upper and lower error bars. The solid line denotes the averaged distance among Zn ions. Shaded area ($y>y_0$) indicates the region in which the elastic IC correlations were observed in neutron scattering measurements (Refs. 12–14). Horizontal dashed line represents the diameter of the nonsuperconducting (non-SC) $2\xi_{nSC}$ estimated by μSR measurements (Ref. 19). (b) Schematic drawing of an inhomogeneous mixture of the superconducting (SC) and non-SC regions for both $y<y_0$ and $y>y_0$. White color area corresponds to the SC region and gray area is the non-SC one.

consistent with the onset temperature in resistivity in the high-T_c cuprates.[17] Thus, it is expected that $T_c(H)$ is located around the onset T_c. In the present analysis, we use $T_c(r=0.95)$ as $T_c(H)$. The critical field at $T=0$ K, $H_{c2}(0)$, is extrapolated using the Werthamer-Helfand-Hohenberg formula, given as[18]

$$H_{c2}(0) \simeq -0.7T_c\left(\frac{dH_{c2}(T)}{dT}\right)_{T=T_c}. \quad (1)$$

Then, ξ is evaluated through the relation $H_{c2}(0)=\phi_0/2\pi\xi_{ab}^2$ for $H\parallel c$, where ϕ_0 is the magnetic flux quantum ($=hc/2e$ $=206\,700$ T Å^2). Figure 4(a) shows the Zn-doping dependence of ξ_{ab} thus estimated. Note that $T_c(H)$ defined from $T_c(r=0.95)$ contains some uncertainties for $y=0-0.008$ showing a broad transition, which are affected by large superconducting fluctuations. This fact would lead to an overestimation of ξ_{ab}. To check the validity of our analysis, we also evaluated the coherence length ξ_{ab} using $T_c(r=0.98)$ and $T_c(r=0.90)$ as $T_c(H)$. The estimated values of ξ_{ab} are shown as error bars in Fig. 4(a). The lower error bars are virtually invisible because ξ_{ab} estimated from $T_c(r=0.98)$ is almost same as that from $T_c(r=0.95)$. We have thus concluded that the uncertainties are small enough to discuss the Zn-doping dependence of ξ_{ab} in further details.

As seen in Fig. 4(a), ξ_{ab} for both ρ_{ab} and ρ_c demonstrate a tendency to increase upon doping Zn. It is remarkable that ξ_{ab} for ρ_{ab} is much longer than that for ρ_c at $y=0.017$, reflecting the parallel shift behavior. Here ξ_{ab} estimated from ρ_{ab} is basically same as that from ρ_c. These results imply that ξ_{ab} for ρ_{ab} drastically changes at the boundaries of y $=0.008$ and $y=0.017$, which we define as y_0. At the boundary y_0, the magnetic property also changes; there is no magnetic static correlation for $y<y_0$, but the IC magnetic peaks appear for $y>y_0$. It indicates that the transport properties in the CuO_2 plane may be related to the two-dimensional magnetic correlations. In $YBa_2Cu_3O_{6.9}$, Tomimoto *et al.*[20] reported that the coherence length grows with increasing Zn and the increase in ξ_c is larger than a theoretical prediction. The rapid increase in ξ_c may give rise to a tendency toward the parallel shift, because the superconductivity comes close to three dimensional. The present results also show that ξ_{ab} gradually increases except for ρ_{ab} for $y=0.017$ and are consistent with their report for Zn-doped $YBa_2Cu_3O_{6.9}$. However, the sudden change in ξ_{ab} for ρ_{ab} at $y=y_0$ requires another scenario.

We offer an inhomogeneous model to explain the sudden growth of ξ_{ab} for ρ_{ab} at $y=y_0$. Our concept is schematically shown in Fig. 4(b). For the Zn-doped system, Nachumi *et al.*[19] proposed an inhomogeneous picture, which they called the "swiss cheese" model; nonsuperconducting (non-SC) islands with the radii ξ_{nSC} around Zn ions reside in the superconducting (SC) sea. Furthermore, scanning tunneling microscopy (STM) studies for Zn-doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ showed that Zn induces an intense quasiparticle-scattering resonance around Zn, indicating that the superconductivity is strongly suppressed within ξ_{nSC}.[21] The radius of an island, ξ_{nSC}, estimated by μSR and STM correspond to 18 Å and 15 Å. Near $y=y_0$, the average distance between Zn ions $r_{\text{Zn-Zn}}$ roughly corresponds to $2\xi_{nSC}$, and the IC magnetic peaks appear for $y>y_0$. From the inhomogeneous picture [see Fig. 4(b)], when $r_{\text{Zn-Zn}}$ reaches $2\xi_{nSC}(y=y_0)$, the isolated nonsuperconducting islands become connected and the IC magnetic correlations can be stabilized. The non-SC islands float in the SC sea for $y<y_0$, while small SC lakes remain in the non-SC land for $y>y_0$. In the case of $y>y_0$, the SC regions are isolated and can be connected through a Josephson tunneling across the non-SC land, which can realize superconductivity. Some results suggest that the superconductivity near the critical concentration, y_c, at which T_c is fully suppressed, is not a bulk property,[22,23] supporting our idea. Why is the superconducting fluctuation suppressed in the region of $y>y_0$? A possible reason can be that the three-dimensional superconductivity is realized in highly Zn-doped region. In fact, Tomimoto *et al.*[20] reported that ξ_c increases upon Zn doping in $YBa_2Cu_3O_{6.9}$. However, such a crossover of dimensionality of superconductivity would not explain why ξ_{ab} of ρ_{ab} is different from that of ρ_c for $y<y_0$. We speculate that ρ_{ab} can be affected by a development of IC correlation in the two-dimensional CuO_2 plane. It is known that the static IC correlation competes with the

superconductivity. The development of IC correlation might interrupt the growth of superconductivity. To address this issue, further studies are required.

Another effect of Zn is that the resistivity exhibits an insulating behavior.[24,25] It has been discussed that the insulating behavior is caused by a large residual scattering[24] or a charge localization,[25] which can destroy the superconductivity in the vicinity of Zn. In the non-SC region around Zn, the static IC magnetic correlations develop, suggesting that the static IC correlation competes with the high-T_c superconductivity, which was suggested by a previous μSR measurement.[26] The parallel shift and the IC static correlation were observed not only in the Zn-doped system, but also in $La_{2-x}(Nd,Sr,Ba)_xCuO_4$ around $x=1/8$,[6–8] as mentioned above. In $La_{2-x}Ba_xCuO_4$ with $x=0.10$, the broadening was observed in low magnetic fields, and it changes to the parallel shift in high magnetic fields accompanied by the insulating behavior of the normal state.[7] This result suggests that the insulating behavior is related to the parallel shift. It is known that the resistivity under an ultrahigh magnetic field exhibits such an insulating behavior in the underdoped regime.[27,28] One may expect that the parallel shift and the static IC correlations are observed under ultrahigh magnetic fields. Further studies are required to clarify the relation among the parallel shift, the insulating behavior of the normal state, and the appearance of static IC correlation in the underdoped regime.

Finally, we briefly discuss the similarity between Zn doping and vortex. Following the swiss cheese model, the non-SC islands are introduced around Zn but the superconductivity in the SC sea remains nearly unaffected by Zn. Therefore, the suppression of T_c by Zn doping mainly originates in the reduction of superfluid density, which is analogous with the case of vortex. Note that the estimated ξ_{ab} for $y=0$, which corresponds to the size of vortex core, is close to the value of ξ_{nSC}. As seen in Figs. 3(a)–3(c), the gradient dH/dT of $r=0.95$ is unchanged for ρ_c for $y=0-0.008$. The gradient displays how much of superconducting area is suppressed by magnetic field, which can reflect characteristic properties of superconductivity. We speculate that the constant gradient indicates little change in superconductivity by Zn, which is consistent with our inhomogeneous picture at $y<y_0$.

V. CONCLUSIONS

We performed electrical resistivity measurements under various magnetic fields for $La_{1.85}Sr_{0.15}Cu_{1-y}Zn_yO_4$ ($y=0,0.004,0.008,0.017$). A typical broadening behavior was observed for $y=0-0.008$, while the resistivity curves in ρ_{ab} at $y=0.017$ exhibit a parallel shift; likewise, at $y=0.017$, the IC static correlation was observed unlike $y=0-0.008$. We conclude that the parallel shift is associated with the static IC correlation in the Zn-doped $La_{1.85}Sr_{0.15}CuO_4$ and that these phenomena are observed not only in the high-T_c materials around the 1/8 hole concentration but also in impurity-doped system. The change from the broadening to the parallel shift suggests that the coherence length ξ_{ab} for $y<y_0$ ($0.008<y_0<0.017$) is much shorter than that for $y>y_0$. It can be explained by an inhomogeneous picture; when the average distance among Zn ions $r_{Zn\text{-}Zn}$ reaches the diameter of the non-SC region $2\xi_{nSC}(y=y_0)$, the non-SC areas connect with each other, while the remaining small SC regions are separated from one another. In the case, the IC correlation in a non-SC area develops and the superconductivity with small superconducting fluctuation is realized in the isolated SC regions.

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