Challenges to high-$T_c$ superconductivity in cuprates by exploring condensate properties

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Abstract.
In this paper, I introduce recent our results on superconductivity fluctuation measurement of high-$T_c$ cuprate both for the hole-doped La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) and the electron-doped La$_{2-x}$Ce$_x$CuO$_4$ (LCCO). In hole doped LSCo, the universality class was found to change twice as a function of doping, starting from the 2D-XY, changing to the 3D XY and another 2D “unknown" behavior. The results favors theoretical interpretations of the phase diagram of high-$T_c$ cuprate which assume the existence of an additional hidden quantum critical point around at the optimum doping. In electron doped cuprate, the superconductivity fluctuation is 3D XY for all samples with different hole doping, which is in sharp contrast to the hole doped cuprate. Thus, the asymmetry of the phase diagram between the hole doped and the electron doped materials is another important key factor to judge the applicability of various theories on high-$T_c$ superconductivity.

Under finite magnetic fields, the scaling relation was valid only for weak fields, and for higher fields, aspects as vortices appeared. However, even at low temperatures, just above the first order phase transition, vortex picture alone cannot describe the data satisfactorily. Thus, we need a unified theory for the description of a large superconductivity fluctuation under finite magnetic fields for high-$T_c$ cuprates.

Finally, I showed that our novel technique of fabricating high-$T_c$ Josephson bridge junction using a small island of Fe was turned out to be very promising.

1. Introduction
High-temperature superconductivity of cuprates has been providing many challenges to condensed matter scientists continuously since its discovery in 1986[1]. Even the pile up of tremendous number of experimental results and theories have not been able to reach the essential understanding of the phenomena, for which all the scientists accept as the ultimate one. One of the reasons for this continuous controversial situation is that many of the experimental approaches were for the normal state of the cuprate superconductors, where the effects of disorder and other effects specific to particular materials might hinder the essential common aspects of the phenomena. From this viewpoint, the investigation of properties of the quantum condensate itself can mask many features which are not important for the essential understanding of the high-$T_c$ superconductivity in cuprates. Thus, we focus on properties in the condensate of cuprate superconductors, and try to put restrictions on the possible theoretical interpretations of the phenomena by investigating condensate properties. Below, we discuss superconductivity fluctuation as a function of doping[2, 3, 4].
Another challenge of high-$T_c$ cuprate superconductors is to look for novel applications. Particularly promising is the single-flux quantum (SFQ) device using high-$T_c$ superconductors[5]. The SFQ device has been expected to act as an ultra-low-power-consumption and ultra-high-speed device, since the power consumption takes place only when a single flux quantum crosses the Josephson junction. Indeed, integrated circuits have been fabricated in conventional superconductors, which work successfully[6]. Since the speed of the SFQ device is proportional to the so-called $I_c R_N$ product, which is, in turn, proportional to the critical temperature, $T_c$[7], high-$T_c$ cuprate superconductors are particularly promising for ultra-high-speed SFQ device. However, even after 20 years of the discovery of the high-$T_c$ superconductivity, Josephson junctions with a satisfactory $I_c R_N$ value are lacking[8]. This is probably due to the very difficult science of interface between the cuprate and the oxide barriers. Thus, we need to develop novel method to fabricate Josephson junction without oxide barriers. Here, I present our recent approach for this task[9].

2. Experiments
Fluctuation in superconductivity was investigated by measuring the ac complex conductivity in the so-called broadband technique[10], where microwave frequency was swept in a continuous manner and the complex reflection from the samples placed at the end of the coaxial cable was measured. For this technique, thin films were used to make the analysis easy. Details of our techniques were described in refs.[2, 3, 4].

Hole doped La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) films were fabricated by the pulsed laser deposition (PLD) method[11], whereas electron doped La$_{2-x}$Ce$_x$CuO$_4$ (LCCO) films were fabricated by the molecular-beam-epitaxy (MBE) method[12].

Josephson junction was fabricated by the electron beam lithography technique. A small island of Fe was put on the superconductor strip of LSCO film (optimally doped) grown on the LSAO substrate. The thickness and the width of the strip are typically 60 nm and 2 μm, respectively, and the thickness and the length of the Fe island are 60 nm and 300 nm, respectively. Details on the process of the fabrication will be published elsewhere[9].

3. Results and discussions
3.1. Superconductivity fluctuation
3.1.1. hole doped cuprates: LSCO Figure 1 shows an example of the zero-field complex conductivity of underdoped LSCO samples as a function of frequency at various temperatures. Both of the real part and the imaginary part increases with decreasing frequency, and the increase becomes more pronounced when temperature approaches $T_c$ from above. This is a clear manifestation of the superconductivity fluctuation. As for the dependence on doping, we previously clarified[2] that in all the underdoped samples ($x=0.07 \sim 0.14$) the superconductivity transition is the Berezinsky-Kosterlitz-Thouless (BKT) type[13], based on the temperature dependence of the superfluid density measured at different frequencies. This feature becomes less clear suddenly in the optimally doped samples. To investigate the origin of the sudden change in the nature of the superconductivity transition, we made the scaling analysis of the complex conductivity data. Scaling analysis assumes that the critical behaviors are characterized by the divergence of typical length- and time scales in the vicinity of a critical point. In the case of the classical phase transition[14], a correlation length, $\xi$, is often used as the typical length scale. $\xi$ diverges as $\xi = \xi_0 |T/T_c - 1|^{-\nu}$, where $\xi_0$ is the correlation length at $T = 0$ K and $\nu$ is a static critical exponent. On the other hand, the divergence of a correlation time, $\tau$, is given by $\tau = \xi^z$. Here, $z$ is a dynamic critical exponent. Fisher, Fisher, and Huse (FFH) [15] provided a general formulation of the dynamic scaling hypothesis for the frequency-dependent complex fluctuation
Figure 1. Complex conductivity, $\sigma = \sigma_1 - i\sigma_2$ of underdoped LSCO ($x=0.07$, 0.12, and 0.14) as a function of frequency at various temperatures.

conductivity, $\sigma_B(\omega)$, near the superconductivity transition, as follows,

$$\sigma_B(\omega) \approx \xi^{2-d} S(\omega \xi^d).$$

Here, $S(x)$ is a complex universal scaling function and $d$ is a spatial dimension. As was emphasized in our previous paper[2], the most essential part of our analyses is that we can check the validity of this hypothesis, by measuring the frequency dependence of $\sigma_B(\omega)$.

Figures 2 and 3 are the scaling plots of the amplitude and the phase of the complex conductivity for underdoped and optimally doped samples. Corresponding to the above mentioned feature, all the underdoped materials can be well expressed by the two dimensional (2D) XY model (temperature dependence of the scaling parameters are not shown here), whereas the optimally doped samples can be expressed by the three dimensional (3D) XY model. The change in the scaling behavior from the underdoped samples to the optimally doped samples takes place very suddenly. The sudden change in the scaling behavior cannot be explained by size effects, as will be discussed briefly later.

In figures 3 and 4, there is another sudden change (3D XY to another 2D behavior) in the scaling behavior when the doping proceeds further. This is highly unexpected, since it has been generally established that cuprate samples become less anisotropic when doping proceeds. Based on the repetition of the same measurements in many samples with various thicknesses, it was found that the change cannot be explained by the size effect, again. Concerning on the
critical exponents, the scaling analysis for the 2 dimension solely provides the product, $\nu z$. The measurement of the mean-field upper critical field as a function of temperature gives $\nu = 0.9$, which provides $z = 1.7$. These exponents do not coincide with any of the well established models, such as 3D XY nor 2D XY models, gaussian models, etc. However, it is interesting to note that many of the extended XY models including randomness, disorders show the exponent $\nu$ of 0.8~$\sim$~1.0[16]. This suggests that the 2D behavior in the overdoped region is closely related to any effects caused by the disorders, and we expect that in overdoped samples with less disorders the BKT transition revives.

Based on these results, we consider that the superconductivity transition of the hole doped cuprate is summarized as follows (Figure 5). In the underdoped samples, it is 2D-XY like (BKT like). With doping, it changes suddenly in to 3D XY like, and changes suddenly again into another 2D like with exponents, $\nu = 0.9$ and $z = 1.7$. In other words, the 3D feature was observed only in a narrow region of the hole doping around optimally doped samples.

This reminds us theoretical explanations where they assume an additional quantum critical point almost at optimal doping. Since the classical 2D thermal fluctuation is mapped onto the quantum 3D fluctuation[17], it may be possible that the large quantum fluctuation around the additional quantum critical point affects the classical thermal fluctuation around the optimal doping, leading to the 3D XY behavior near optimal doping.
Figure 3. Scaling plot for the amplitude and the phase of complex conductivity of optimally doped LSCO samples with various doping.

Figure 4. Scaling plot for the amplitude and the phase of complex conductivity of overdoped LSCO samples with various doping.
3.1.2. **electron doped cuprates; LCCO** In electron doped cuprates, LCCO, all samples we measured show superconductivity fluctuation with the 3D XY feature, independent of doping. This is shown schematically also in figure 5. This result is in sharp contrast to the hole doped cuprate. This suggests that any theoretical interpretations of high-$T_c$ superconductivity assuming the symmetry between the hole-doped and the electron-doped materials are inappropriate.

3.2. **Superconductivity fluctuation under finite magnetic fields**

High-$T_c$ cuprates show a characteristic phenomenon called “resistive broadening” under finite magnetic fields[18, 19]. The significance of the phenomena is that the conventional upper critical field, $B_{c2}$, only has the meaning in the mean-field sense. The true phase transition takes place at much lower fields, which is considered to be the first order vortex-solid-liquid transition[19]. Thus, the vortex liquid state is the same as the normal state in the thermodynamical sense. However, just above the transition, the vortex picture is still appropriate. Therefore, it is very interesting how the vortex picture crosses over to the ordinary fluctuation picture with increasing temperature/magnetic field.

3.2.1. **weak magnetic fields** For low fields (typically up to 0.1 T), ac conductivity data can be analyzed exactly by the same method as was made in zero magnetic field. For underdoped LSCO, it was found that the exponential divergence characteristic of the 2D-XY BKT feature was suppressed by introducing external magnetic flux. On the other hand, for all other samples, the scaling relation hold rather well even in finite magnetic fields. From this result, one can estimate the upper limit of the in-plane correlation length of the fluctuation, $\xi_{ab}$. This, in turn,
provides the upper limit of the interplane correlation length of the fluctuation $\xi_c$, since $\xi_c < \xi_{ab}$. It turned out that the estimated interplane correlation length $\xi_{ab}$ is smaller than the thickness of the film. Thus, it became clear that the 2 dimensional feature observed in the overdoped region was not caused by the size effect, namely, the competition between the sample thickness and the interplane correlation length.

3.2.2. high magnetic fields The scaling relation did not hold for the data taken at higher magnetic fields. For these data, we tried to analyze the data in terms of the following formula,

$$\rho \equiv \rho_1 - i\rho_2 = \frac{\rho_{\text{vortex}} + i\mu_0\omega\lambda^2}{1 + 2i\lambda^2/\delta_{nf}^2} \simeq \rho_{\text{vortex}} + i\mu_0\omega\lambda^2 \quad (\delta_{nf} \gg \lambda),$$

(2)

where $\rho \equiv \rho_1 - i\rho_2$, $\rho_{\text{vortex}}$ are the complex resistivity, and the resistivity by the vortex motion, respectively, $\mu_0$, $\omega$ are the permeability of vacuum, angular frequency, respectively, and

$$\delta_{nf} = \sqrt{\frac{2\rho_0}{\mu_0\omega}}$$

(3)

is the normal fluid skin depth ($\rho_0$ is the resistivity in the normal state), and $\lambda \equiv \sqrt{m/n_s e^2}$ is the London penetration depth ($n_s$, $m$ and $e$ are the superfluid density, mass, and charge of superconducting electron). As for the vortex part, we used the expression derived by Coffey and Clem based on the mean-field treatment of the vortex motion[20],

$$\rho_{\text{vortex}} = \left( \frac{B\Phi_0}{\eta} \right) \left[ \varepsilon + \frac{(\omega\tau_p)^2 + i(1 - \varepsilon)\omega\varepsilon\tau_p}{1 + (\omega\tau_p)^2} \right],$$

(4)

where $\eta$ and $\tau_p^{-1}$ are the viscosity and the crossover frequency of the vortex, respectively, $\varepsilon$ is the flux-creep factor, $B$ and $\Phi_0$ are the magnetic field and the flux quantum, respectively. We regard $\eta$, $\tau_p$, $\varepsilon$, and $\lambda$ as fitting parameters, and obtained them by fitting the measured $\rho$ data to the above formula. Figure 6 shows the parameters obtained by these analyses. Most of these results show that the obtained parameters are reasonable, and the vortex picture holds rather well. However, the penetration depth decreases with increasing temperature, which is rather unusual. This suggests that the vortex picture alone is insufficient to explain the experimental data of ac conductivity under finite magnetic fields in this region. Thus, even at rather low temperatures, the conventional picture of vortex motion collapsed. Unfortunately, we do not have any theoretical formula for these crossover regions, and a unified treatment of the large superconductivity fluctuation in cuprates is needed urgently.

3.3. Fabrication of Josephson junction by a novel method

As was already mentioned in section 1, the fabrication of good Josephson junctions without any extra oxide barrier layers is needed for practical use of cuprate Josephson junction. We fabricated a novel bridge type junction, schematically shown in Figure 7, where a small island of magnetic Fe is placed on the top of a small high-$T_c$ strip. The magnetism of Fe is expected to destroy superconductivity below (probably because of the exchange coupling, not the magnetic coupling), and Josephson junction might be fabricated. Figure 8 shows the $I - V$ characteristics of the junction at several different temperatures. For comparison, the $I - V$ characteristics of a junction with a Cu island, instead of Fe, was also shown. The $I - V$ characteristics of the Cu junction is typical of the flux-flow type, whereas those of the Fe junction shows concave upward behavior in the finite voltage state, which is indicative of the formation of weak links. Although this is rather different from the textbook like $I - V$ characteristics of the Josephson junction, we
Figure 6. Temperature dependences of parameters for data in high-magnetic fields of LCCO obtained by the ac conductivity measurements.

tentatively define the $R_N$ value as the chordal resistance of the maximum voltage applied. Thus, we obtain the $I_c R_N$ value of about 10 mV, which is one of the largest value ever obtained for cuprate superconductors[21]. To show that the fabricated structure is the Josephson junction more clearly, we investigated the $I_c$ as a function of magnetic field and the $I - V$ characteristics under finite microwave radiation both for the Fe sample and the Cu sample. As for the $I_c$ as a function of magnetic field, Cu samples did not show any appreciable change as a function of magnetic field, whereas the $I_c$ of the Fe sample shows a broad bell shaped behavior, reminiscent of the principal peak of the Fraunhofer pattern. Figure 9 shows the $I - V$ characteristics of the Fe sample with and without the 2 GHz microwave radiation. The data with the microwave radiation shows a clear interference structure, which is exactly the Shapiro step, whereas the Cu sample merely shows the decrease of $I_c$ without any interference structure (not shown here). In addition, we investigated the step height as a function of the amplitude of microwave radiation, and obtained the result which is typically expected for the Shapiro steps[9]. All of these data show that the Fe sample becomes a Josephson junction. For the SFQ use, it is preferable to demonstrate that a single flux quantum moves crossing the Josephson junction, indeed. Thus, we fabricated a dc SQUID by this method and measured the voltage as function of magnetic field, the result of which is shown in figure 10. Definite oscillations were observed with a period exactly corresponding to one flux quantum. Therefore, our novel method presented here is very promising for the application of the high-$T_c$ Josephson junction to the SFQ device.
4. Conclusion

In this paper, I introduced recent our results on superconductivity fluctuation measurement of high-$T_c$ cuprate both for the hole doped LSCO and the electron doped LCCO. In hole doped LSCO, the universality class was found to change twice as a function of doping, starting from the 2D-XY, changing to the 3D XY and another 2D “unknown” behavior. The results favors theoretical interpretations of the phase diagram of high-$T_c$ cuprate which assume the existence of an additional hidden quantum critical point around at the optimum doping. In electron doped cuprate, LCCO, the superconductivity fluctuation is 3D XY for all samples with different hole doping, which is in sharp contrast to the hole doped cuprate. Thus, the asymmetry of the
Figure 9. $I - V$ characteristics of the Fe junction with and without the 2 GHz microwave radiation. To highlight the interference structure, dV/dI was also shown for the data with the microwave radiation.

Figure 10. Voltage of the dc SQUID fabricated using Fe junctions as a function of magnetic field.

Phase diagram between the hole doped and the electron doped materials is another important key factor to judge the applicability of various theories on high-$T_c$ superconductivity.

Under finite magnetic fields, the scaling relation was valid only for weak fields, and for higher fields, aspects as vortices appeared. However, even at low temperatures, just above the first order phase transition, vortex picture alone cannot describe the data satisfactory. Thus, we need a unified theory for the description of a large superconductivity fluctuation under finite magnetic fields for high-$T_c$ cuprates.

Finally, I showed that our novel technique of fabricating high-$T_c$ Josephson bridge junction using a small island of Fe was turned out to be very promising for the application to the SFQ device.

Acknowledgment
This work is in collaboration with T. Ohashi, H. Kitano, I. Tsukada, M. Naito, A. Tsukada, L. Gomez and S. Kitamura. I thank all of these people for collaboration.
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