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Out-of-plane dielectric constant and insulator–superconductor transition in $Bi_2Sr_2Dy_{1-x}Er_xCu_2O_8$ single crystals

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Abstract

The out-of-plane dielectric constant of the parent insulator of the high-temperature superconductor $Bi_2Sr_2Dy_{1-x}Er_xCu_2O_8$ was measured and analysed from 80 to 300 K in the frequency range of 10^6–10^9 Hz. All the samples were found to show a fairly large value of 10–60, implying some kind of charge inhomogeneity in the CuO_2 plane. Considering that the superconducting sample $Bi_2Sr_2Ca_{0.75}Pr_{0.25}Cu_2O_8$ also shows a similar dielectric constant, the charge inhomogeneity plays an important role in the insulator–superconductor transition.

1. Introduction

A cuprate with a nearly undoped CuO_2 plane shows high resistivity and antiferromagnetic order at low temperature, which is called a parent insulator of high-temperature superconductors (HTSC). With doping, an insulator–metal transition (IMT) arises, and the system changes from the parent insulator to HTSC. For IMT, the dielectric constant ε is of great importance in the sense that it provides a measure of localization length in the insulator. In particular, ε for doped Si divergingly increases with a critical exponent toward the IMT [1].

Chen *et al* [2] first pointed out the importance of ε for the IMT of HTSC. They found that ε of $La_2CuO_{4+\delta}$ (La-214) diverged only along the in-plane direction toward the IMT, and stated that the IMT of HTSC was two-dimensional. However, their study was done only for La-214, in which a quasi-static stripe fluctuates in the CuO_2 plane [3]. Since the stripe fluctuation is a kind of charge modulation and/or a charge disproportion, it might cause a large dielectric response. Perhaps related to this, La-214 shows structure phase transition [4], oxygen ordering [5] and phase separation [6], which might also give a large dielectric response. Thus it is necessary to study another class of HTSC, which is far away from the stripe- or lattice-instability.

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We think that $Bi_2Sr_2RCu_2O_8$ (Bi-2212) is the best candidate. This class of HTSC shows no orthorhombic–tetragonal (OT) phase transition below 300 K, and is far away from the instability as is seen in the overdoped La-214 [4] in which T_c and the OT transition temperature are very close. We further expect that Bi-2212 is away from the stripe-instability in the sense that the well-defined cold spot is observed near $(\pi/2, \pi/2)$ in the photoemission spectra [7, 8]. Note that the photoemission spectra of underdoped La-214 have no cold spot (or, equivalently, no Fermi surface) near $(\pi/2, \pi/2)$ [9], which is considered to be a piece of evidence for the quasi-static stripes, because the holes on the stripes along the [100] direction cannot move along the [110] direction. Of course we will not say that Bi-2212 is an ideal cuprate; some researchers have reported its lattice-instability [10] and its stripe-instability [11–13]. What we would like to emphasize is that these instabilities would be smaller for Bi-2212 than for La-214, which is also suggested from a theoretical viewpoint by Tohyama and Maekawa [14]. Thus Bi-2212 can be a counterpart to La-214, which will help our understanding of the dielectric response of HTSC themselves.

We have studied the charge transport of the parent insulator of Bi-2212 [15–17], and have found that the physics is as rich as the physics of HTSC themselves. In this paper, we report on measurement and analysis of the dielectric constant of $Bi_2Sr_2Er_{1-x}Dy_xCu_2O_8$ single crystals along the out-of-plane direction.

2. Experimental

Single crystals of Bi-2212 were grown by the self-flux method. The growth conditions and the sample characterization were described in [15]. The prepared crystals of $Bi_2Sr_2Dy_{1-x}Er_xCu_2O_8$ ($x = 0$, 0.3, 0.5, 0.7 and 1.0) were insulating, and the doping levels of the as-grown crystals were slightly different for different x. An energy-dispersive x-ray (EDX) analysis revealed that Sr was slightly excessive, and the content was estimated to be $Bi_{2.2}Sr_{3.4-y}R_yCu_2O_{8+\delta}$ (within an experimental error of 10%). The R content y was 1.0 for R = Dy and 0.7 for R = Er. Thus a real composition would be something like $Bi_2Sr_2DyCu_2O_8$ and $Bi_2Sr_2(Sr_{0.3}Er_{0.7})Cu_2O_8$ for R = Dy and Er, respectively. Thus a solid solution between Er and Dy gives $Bi_2Sr_2Dy_{1-x}(Sr_{0.3}Er_{0.7})_xCu_2O_8$, which enables us to finely tune the doping level from a highly insulating ($x = 0$) to a slightly doped level ($x = 1$). Instead of the real composition, we will call the prepared samples $Bi_2Sr_2Dy_{1-x}Er_xCu_2O_8$ as a matter of convenience. For reference, a superconducting sample $Bi_2Sr_2Ca_{0.75}Pr_{0.25}Cu_2O_8$ ($R = Ca_{0.75}Pr_{0.25}$) was also prepared.

Figure 1(*a*) shows a photograph of a prepared sample of $x = 0$. The surface is shiny and smooth with no serious cracks. Figure 1(*b*) shows a Laue pattern of the *ab* plane of the crystal. The quasi-four-fold symmetry is observed, although the spots are somewhat diffusive because the sample was thin and slightly warped. The modulation structure is clearly observed in the TEM diffraction image as shown in figure 1(*c*). The sample surface is perpendicular to the [001] direction, which is evidenced by the fact that the x-ray diffraction pattern showed only the (00 2ℓ) reflections, as shown in figure 1(*d*). The Er substituted samples of $x \neq 0$ showed essentially similar results, although the sample size of $x = 0.3$, 0.5 and 0.7 was somewhat smaller than that of $x = 0$ and 1. These data indicate that the crystal quality is as good as that for single crystals of superconducting Bi-2212 prepared by the flux technique.

The complex impedance of the samples in the frequency range of 10^6–10^9 Hz was measured with a two-probe technique using an rf LCR meter (Agilent 4287A) using a similar technique to Böhmer *et al* [18]. A low contact resistance was realized by uniformly painting the silver paste (Dupont 6838) on both sides of the c-plane surface, followed by annealing at 873 K for 30 min. The contact resistance was 1–2 Ω, which can be safely neglected by comparison

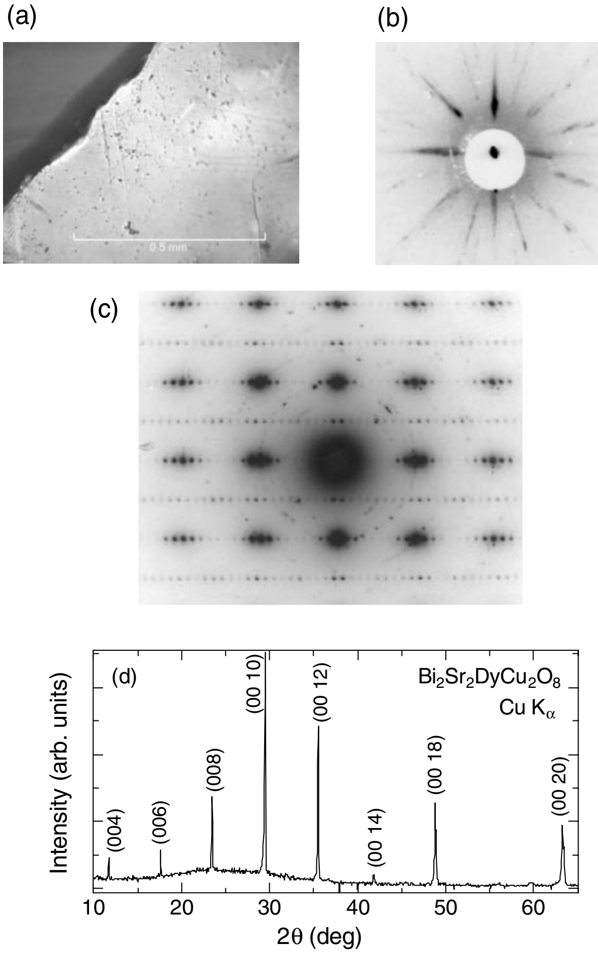

Figure 1. (*a*) Photo picture, (*b*) Laue photo image, (*c*) TEM diffraction pattern and (*d*) x-ray diffraction pattern in the $\theta - 2\theta$ scan mode of a single-crystal sample of $Bi_2Sr_2DyCu_2O_8$.

with the sample resistance (typically 100 Ω). In order to make the sample resistance high, we cut the sample along the a and b directions as small as possible. For the most conducting sample of R $=$ $Ca_{0.75}Pr_{0.25}$, the dimension was $0.2 \times 0.2 \times 0.05$ mm^3. Figure 2(*a*) shows a typical two-probe resistance for R $=$ $Ca_{0.75}Pr_{0.25}$. Since the sample is superconducting, the resistivity rapidly decreases below 80 K, and the residual resistance is 6.6 Ω which is the sum of the contact resistance and the probe resistance (∼5 Ω). The sample resistance is 280 Ω at 80 K, which indicates that the contribution of the contact resistance is less than 1%.

As is schematically shown in figure 2(*b*), the measured impedance was understood as a parallel circuit consisting of a resistance $R(\omega)$ and a capacitance $C(\omega)$ which can be dependent on frequency $\omega = 2\pi f$. Then the complex impedance Z is written as:

$$Z = \frac{R}{1 + \mathrm{i}\omega C R} = \frac{R}{1 + (\omega C R)^2} - \mathrm{i}\frac{\omega C R^2}{1 + (\omega C R)^2}. \qquad (1)$$

The capacitance and resistance are expressed in terms of the dielectric constant ε and the (ac) conductivity σ as $C = \varepsilon_0 \varepsilon A/d$ and $R = d/A\sigma$, where A and d are the area and the distance

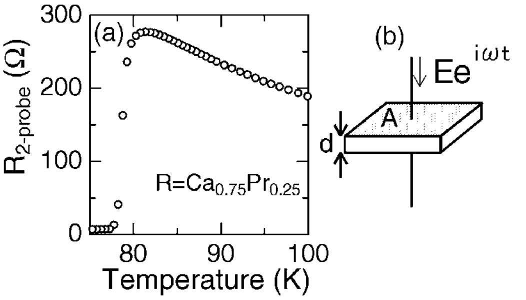

Figure 2. (*a*) The two-probe resistance ($R_{2\text{-probe}}$) for a superconducting sample $Bi_2Sr_2Ca_{0.75}Pr_{0.25}Cu_2O_8$, and (*b*) a schematic drawing for the complex impedance measurement.

of the capacitor, and ε_0 is the dielectric constant of a vacuum. Then ε and σ are obtained from the complex impedance as:

$$\varepsilon = -\frac{d}{\omega\varepsilon_0 A}\frac{\operatorname{Im} Z}{|Z|^2} \tag{2}$$

$$\sigma = \frac{d}{A}\frac{\operatorname{Re} Z}{|Z|^2}. \tag{3}$$

3. Results

Figure 3(*a*) shows the temperature dependence of the out-of-plane resistivity for the measured samples. Reflecting that the carrier concentration increases with increasing Er content, the magnitude systematically decreases with x. For R = $Ca_{0.75}Pr_{0.25}$, the superconducting transition is observed slightly below 80 K.

The in-plane thermopower of the prepared crystals is shown in figure 3(*b*). We estimated the hole concentration per Cu (p) from the room-temperature thermopower [19], and found p = 0, 0.01, 0.015, 0.03, and 0.035 for x = 0, 0.3, 0.5, 0.7, and 1.0, respectively. For R = $Ca_{0.75}Pr_{0.25}$, p was estimated to be 0.14 (underdoped region). The uncertainty in p was typically ±0.005 arising from the scattered data in [19], within which the estimated p is consistent with the p estimated from the Hall coefficient [20].

Figure 4 shows the out-of-plane dielectric constant (ε_c). The magnitude of ε_c reaches a fairly large value of 10–60. Previously we found that the frequency dependence of ε_c for $x = 0$ is explained with the Debye model of dielectric relaxation [21, 22]. As shown in the inset in figure 4(*a*) taken from [21], ε_c is large and constant at low frequencies, and shows a step-like decrease above a certain frequency to reach a small constant value. This is what we expect in the dielectric relaxation. Sekhar *et al* [23] measured ε of the polycrystalline samples of $Bi_2Sr_2RCu_2O_8$ (R = Sm and Y), and found that ε is as large as 10^4 at 300 K, and shows a relaxation behaviour. Lunkenheimer *et al* [24] measured the anisotropic dielectric constant of single-crystal La_2CuO_4, and found the relaxation behaviour along the c-axis direction. For other parent insulators, Shi [25] measured ε of polycrystalline samples of $(La, Gd)_2CuO_4$, and Mazzara *et al* [26] measured ε_c of single-crystal $PrBa_2Cu_3O_7$. These materials also show a large ε with a relaxation behaviour, and we conclude that this is generic in the parent insulator of HTSC.

It is quite difficult to accurately measure ε_c for a superconducting sample. An upturn above 10^7 Hz for R = $Ca_{0.75}Pr_{0.25}$ is an artifact due to an inductance contribution from the

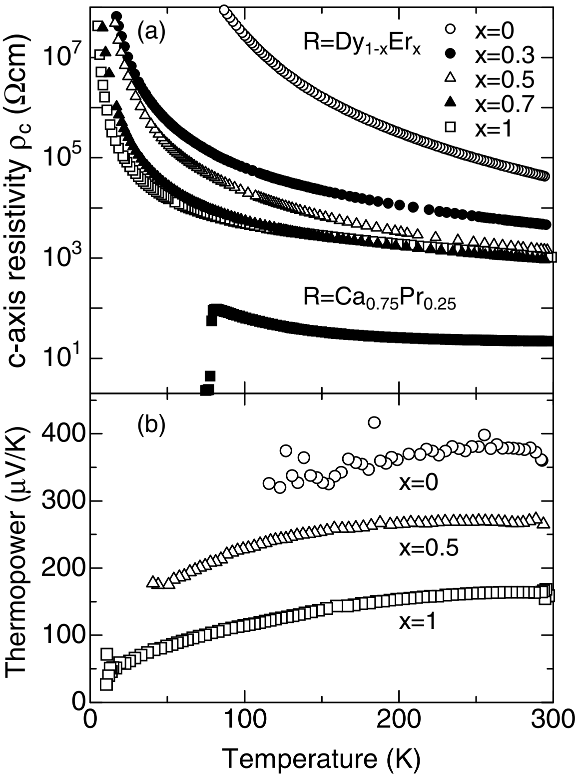

Figure 3. (*a*) The out-of-plane resistivity of $Bi_2Sr_2RCu_2O_8$ (R = $Dy_{1-x}Er_x$ and $Ca_{0.75}Pr_{0.25}$) measured using a two-probe technique, and (*b*) the in-plane thermopower of $Bi_2Sr_2RCu_2O_8$ (R = $Dy_{1-x}Er_x$).

leads because of the small sample resistivity, and an intrinsic dielectric response is likely to be independent of frequency. The low-frequency data is also difficult to measure. Equation (1) requires that ωCR^2 should be in the measurable range of the analyser (typically larger than 1 Ω) but ωCR^2 for R = $Ca_{0.75}Pr_{0.25}$ was too small near 10^6 Hz. A similar situation happened in the samples of $x = 0.7$ and 1 below 10^6 Hz, where we failed to measure ε_c.

An important finding is that ε_c remains positive even in the superconducting sample of R = $Ca_{0.75}Pr_{0.25}$. Kitano *et al* [27] reported nearly the same ε_c of $Bi_2Sr_2CaCu_2O_8$ (40 at 100 K at 10 GHz). This implies no divergence of ε_c at IMT, as Chen *et al* [2] suggested previously. We further note that the out-of-plane charge response is not Drude-like, in the sense that ε is negative below the plasma frequency in the Drude model.

4. Discussion

Let us discuss the magnitude and the frequency dependence of ε_c. First, the large ε_c is not due to phonons or lattice instability in a simple sense, because the large dielectric constant with relaxation behaviour is commonly seen in various parent insulators of HTSC, whose lattice properties are different for a different class of HTSC. Second, then, it should be attributed to an electronic excitation in the CuO_2 plane that is the only common part of the parent insulators. We measured ε_c for the layered cobaltate Bi–Sr–Co–O that has a similar Bi_2O_2 layer, and

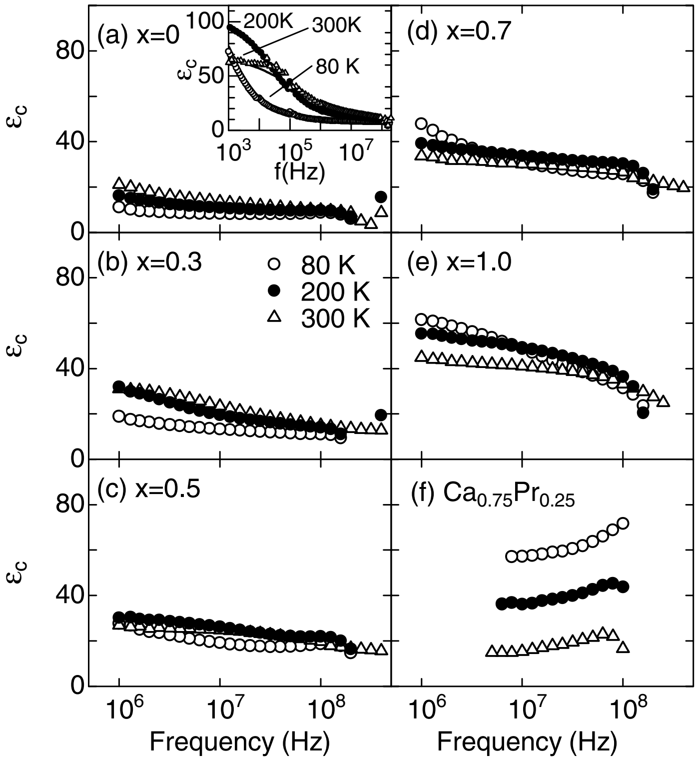

Figure 4. The out-of-plane dielectric constants (ε_c) of $Bi_2Sr_2RCu_2O_8$ ($R = Dy_{1-x}Er_x$ and $Ca_{0.75}Pr_{0.25}$). The inset shows ε_c for $x = 0$ at lower frequencies.

found a small ε_c with essentially no relaxation behaviour [28]. This indicates that neither the other block layers nor the layered structure can be an origin for the large ε_c.

Let us consider the electronic excitation through a simple Lorentz model written as

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{f}{\omega_0^2 - \omega^2 + i\gamma\omega} \tag{4}$$

where f, ω_0, γ are the oscillator strength, the resonance frequency, and the damping rate, respectively. For an overdamped case $\gamma \gg \omega_0$, equation (4) reduces to the Debye model of dielectric relaxation in the low frequency limit $\omega_0 \gg \omega$ as

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{f}{\omega_0^2}\,\frac{1}{1 + i\omega\tau} \tag{5}$$

where $\tau = \gamma/\omega_0^2$. Accordingly a large ε implies a small ω_0 accompanied by a large f. We can demonstrate that ε is small for a simple band insulator, where ω_0 and f correspond to the band gap and the Drude weight of the valence electron. Since these two are of the same order, the second term of equation (5) is of the order of unity for $\omega \to 0$. Note that ε is also small for a Mott insulator, because the Mott–Hubbard gap is of the same order as the band gap. In other words, a single-particle gap is too large to give a large ε.

A collective-excitation gap is most likely to be responsible for a large ε. It is known that the charge-density-wave (or charge-ordered) materials, such as $K_{0.3}MoO_3$ [29], $LuFe_2O_4$ [30], (Pr, Ca)MnO_3 [31] exhibit a large ε with relaxation behaviour, where ω_0 is the pinning frequency of the charge density. This implies that a charge density modulation or a

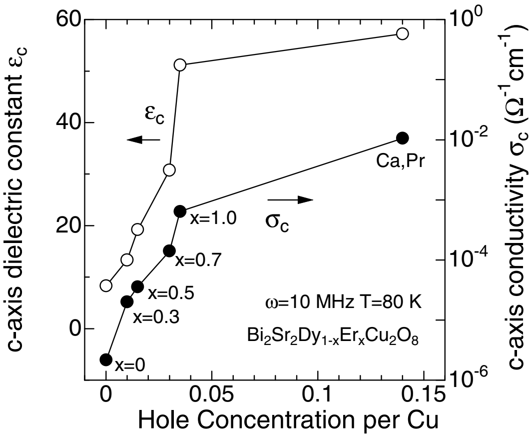

Figure 5. The out-of-plane dielectric constant and conductivity for $Bi_2Sr_2RCu_2O_8$ at 10 MHz plotted as a function of hole concentration per Cu (p). The p was estimated by measuring the room-temperature thermopower.

charge fluctuation exists in the CuO_2 plane, which, in principle, can occur in the doped antiferromagnet. Unlike the case of a doped semiconductor, holes in the parent insulator do not distribute uniformly. They tend to condense in order that they should minimize the exchange-energy loss of the antiferromagnetic background. Prime examples are the charge stripe [3] and the phase separation [32]. Thus we think that the observed dielectric response is a piece of evidence for the charge modulation in the parent insulator. Although phonons may contribute to the charge modulation in the conventional CDW materials, we should emphasize that phonons are not necessarily essential. Nad *et al* [33] have found that the Coulomb repulsion can cause a charge ordered state and a huge dielectric response in an organic material, and we think that the present observation is also the case.

Recent scanning tunnel microscope spectroscopy has revealed unexpected inhomogeneous electronic states of HTSC. Pan *et al* [34] found that the local density of states is inhomogeneous in the CuO_2 plane of the optimally doped Bi-2212, where the doped carriers form a metallic patch. Very recently Howald *et al* [35] also found superconducting and non-superconducting phases (at a nanometer scale) coexist in the CuO_2 plane of the underdoped Bi-2212. This type of charge inhomogeneity could cause large ε, and is consistent with our observations. Note that the inhomogeneity suggested here is seen in a high-quality sample of (nominally stoichiometric) Bi-2212. Also in our case, nominally stoichiometric $x = 0$ and 1 samples show large ε_c, and the solid solution of Er and Dy seem to have little effect on the systematic change in ε_c, as shown in figure 4. The inhomogeneity is something 'intrinsic', and possibly occurring at a nanoscale, which is hard to detect with conventional chemical characterizations. In fact, we did not observe any evidence for 'chemical' inhomogeneity.

Next let us discuss the carrier concentration dependence. In figure 5, ε_c and the out-of-plane conductivity σ_c for a representative frequency of 10^7 Hz at 80 K are plotted as a function of p. Most importantly, finite values of ε_c and σ_c are observed simultaneously. This contrasts strikingly with the case of a doped semiconductor in which the insulating region is characterized by a finite ε and $\sigma = 0$ [1], whereas the metallic region is characterized by a finite σ and $\varepsilon \ll 0$ [36]. Finite ε and σ are simultaneously observed in a highly disordered conductor, where variable-range hopping (VRH) dominates the charge transport [2]. However, in the VRH regime, ε does not show relaxation behaviour, and thus this is not the present case.

The p dependence in figure 5 suggests that the parent insulator is a mixture of insulating and metallic phases. The increase of σ_c with p suggests that carrier-doping makes locally metallic patches in the insulating background. The patches cause a large ε_c by fluctuating its position and a finite σ_c through the proximity effects. Then the charge dynamics can be understood in terms of a percolative mixture of the insulating and metallic phases. Accordingly the insulator–superconductor transition in Bi-2212 occurs when the metallic (superconducting) patches are connected with one another over a whole sample with proximity effects.

Now we will consider the anisotropy of ε. Based on the inhomogeneity scenario, ε is enhanced to be $\varepsilon_\infty + f/\omega_0^2$ for $\omega \to 0$, where f is the Drude weight of the metallic patch that is inversely proportional to the effective mass. It is therefore expected that ε_{ab} is larger by the effective-mass ratio (10^4) than ε_c, whereas the frequency and temperature dependences are nearly identical. This is what we observed in the preliminary measurements for ε_{ab} of R = Dy and Er [22]. In a superconducting sample, the metallic patches are connected from edge to edge, and the carriers can move freely along the in-plane direction. For Bi-2212, carriers near the cold spot $(\pi/2, \pi/2)$ dominate the in-plane transport [8], and are little affected by the inhomogeneity, because the local density of states is rather homogeneous along the $(\pi/2, \pi/2)$ (nodal) direction [34]. This means that a Drude-like charge response can occur along the in-plane direction. Then ε_{ab} changes with doping from positive to negative across the IMT, at which the divergence of ε_{ab} would be seen. On the other hand, owing to the zero matrix element [37], carriers near $(\pi/2, \pi/2)$ cannot move along the out-of-plane direction, and consequently cannot screen the inhomogeneity in the CuO_2 plane. This is the reason the dielectric response along the out-of-plane direction remains unchanged at IMT.

Here we would like to mention existing theories based on the intrinsic inhomogeneity. Just after the discovery of high-temperature superconductivity in 1986, Phillips [38–40] pointed out that the percolation and inhomogeneity should play an important role in HTSC. In his theory, zig-zag filamentary conduction paths are formed near the IMT boundary, where the electron–phonon interaction is highly enhanced to cause high-temperature superconductivity. Chakravarty and Kivelson [41] have found that a nanoscale Hubbard ring with moderate Coulomb repulsion can gain anomalously large pair-binding energy. Burgy *et al* [42] have reported that the intrinsic inhomogeneity causes colossal effects. They have found a close similarity between colossal magnetoresistive manganites and HTSC. All these theories may predict a large ε with relaxation behaviour, and we cannot specify a clear answer among them at present. Nonetheless we hope that ε can be a good probe for the inhomogeneity fluctuation, because it provides us with a lot of valuable information through the frequency, temperature and doping dependences. We hope that a quantitative analysis for ε will specify the nature of the charge inhomogeneity in HTSC.

Finally we will compare our results with those by Chen *et al* [2]. At a qualitative level, our observations are similar to theirs: (1) ε_c increases with decreasing frequency, and (2) ε_c does not diverge at IMT. However our measurement is done for Bi-2212, a counterpart to La-214, including the data for a superconductor. Thus we have successfully shown that the items (1), (2) are generic for HTSC. Another discrepancy is that we understood the frequency dependence as the dielectric relaxation, while they employed VRH. Considering ε of other parent insulators, we think that the relaxation behaviour is generic. Perhaps this comes from the different temperatures measured. They discussed ε at 4 K, where the resistivity is also described in terms of VRH. We found that the resistivity anisotropy is strongly dependent on temperature [15]. The resistivity is almost isotropic at 4 K, and the parent insulator behaves three-dimensionally. Conversely, the resistivity is largely anisotropic above 80 K, and, in particular, the ‘confinement’ is observed even in the parent insulator. Thus it would be natural that the frequency dependence of ε differs between their results and ours.

5. Summary

We prepared single crystals of $Bi_2Sr_2Dy_{1-x}Er_xCu_2O_8$, and measured the out-of-plane dielectric constants in the temperature range of 80–300 K and in the frequency range of 10^6–10^9 Hz. The dielectric constant of the parent insulator is characterized by the large magnitude and relaxation behaviour, which would come from the charge inhomogeneity recently observed in the scanning tunnel microscope experiments. The doping dependence of the dielectric constant and the conductivity suggests that the charge dynamics is understood as a percolation of the insulating and metallic phases, and that the insulator–superconductor transition happens when the metallic (superconducting) patches are connected with one another over a whole sample.

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