

## SI Transitions in BSCCO Mixed Crystal Thin Films

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Temperature ( $T$ ) dependence of the sheet resistance ( $R_{\square}$ ) has been investigated on the  $c$ -axis oriented thin films of the (Bi2212/Bi2201) mixed crystal with different molar fractions. The  $R_{\square}$ - $T$  superconducting characteristic deteriorated with reduction of the Bi2212 fraction, and almost disappears at 48 mol% where a superconductor-to-insulator transition took place, with the resistance on the normal state,  $R_N$ , reaching 4.1 k $\Omega$  at 80 K. This  $R_N$  value is close to the universal quantum number,  $h/(2e)^2 \cong 6.5$  k $\Omega$  predicted by the Kosterlitz-Thouless (KT) transition theory. The  $R_{\square}$ - $T$  characteristics of the 48 mol% thin film can be elucidated as a competitive process of KT transition brought about by charge or vortex in the two-dimensional layer structure.

*Keywords* : SI transition , BSCCO, Mixed crystal, Thin film, KT transition

### 1. INTRODUCTION

It is known that the superconductor-insulator (SI) transition takes place in the two-dimensional array of granular junctions which are in order of submicron size, for instance, Al granular thin film[1]. In such granular films, a key factor for the discrimination between the superconductor and insulator is the critical sheet resistance defined by  $h/(2e)^2 \cong 6.5$  k $\Omega$ , the so-called quantum resistance  $R_q$  which is independent of the materials. Here,  $h$  and  $e$  are the Planck constant and charge, respectively. In the case of the granular junction exhibiting lower resistivity than  $R_q$  in the normal state, the superconductivity comes out with reduction of temperature, while in the one higher than  $R_q$  the resistivity enhances up with reduction of temperature, which results in an insulator.

In the  $\text{Bi}_2\text{Sr}_2\text{Ca}_n\text{Cu}_{n+1}\text{O}_y$  ( $n = 0, 1, \text{ and } 2$ ) compound, which is hereafter abbreviated as the BSCCO or Bi22 $n$ +1), there are three types of the energetically stable structures, that is, the Bi2201 (insulator to superconductor with  $T_c \leq 20$  K), the Bi2212 ( $T_c \cong 80$  K) and the Bi2223 ( $T_c \cong 110$  K), with all of them having a strong two-dimensionality in the structural and electronic states. The difference among these phases

structurally originates in only the introduction of [CaCuO<sub>2</sub>]-stacking fault, so that it is hardly possible to obtain each, pure BSCCO[2]. The mixed crystal thin films prepared in this study are consisted of the Bi2212 (the lattice parameter of  $c$ -axis: about 30 Å) and Bi2201 (about 24 Å) phases with various ratios. Scanning electron microscope observation displays that granular domain size in these thin films does not exceed  $\sim 1 \mu\text{m}$  in a similar condition to the granular films reported by Kobayashi *et al.*[1]. This suggests that the SI transition model in the two-dimensional mixed crystal thin films will be applicable to the thin films with the mixture of the two Bi2201 and Bi2212 phases. Since the resistivity produced depends largely on the arrangement of each phase, even if the grains of the Bi2212 superconducting phase and the Bi2201 insulating one are distributed randomly in the mixed crystal specimen, the electronic current would flow through only the Bi2212 phase channels. The BSCCO thin film must be considered the size effect in atomic scale as well as in the mesoscopic one, differ from a granular Al thin film because the supercurrent flows in only the [CuO<sub>2</sub>]-layer in the BSCCO and coherence length,  $\xi$ , is much shorter than that of Al. Accordingly, in cases that the Bi2212 phases

parated by the intermediate Bi2201 phase completely, the SI transition and the zero resistance would happen. Namely, it will be possible to discuss these resistivities without exact knowledge concerning the inside of the domain size in the BSCCO thin films.

In this article, we tried to discuss about the appearance of the SI transition in the BSCCO mixed crystal thin films from the comparison between the normal state resistance,  $R_{\square}$ , in the normal state and  $R_{q,0}$ . Moreover, temperature dependence of  $R_{\square}$ , will be elucidated by a competitive process of vortex and/or large Kosterlitz-Thouless (KT) transitions proposed by Fazio and Schön[3].

## 2. EXPERIMENTAL

The mixed crystal thin films with various molar fraction ratios between the Bi2212 ( $T_c(\text{onset}) = 82$  K) and the Bi2201 (insulator) phases have been fabricated by an ion beam sputtering method[4]. The measurement of X-ray diffraction patterns on these mixed crystal samples displayed that a series of the respective peaks come from the (00 $l$ ) reflections appeared as single peaks at the intermediate positions between the respective peaks from the pure Bi2212 and Bi2201 substances. This fact indicates that the grain sizes in the mixed crystals are enough smaller than the X-ray penetration length of micrometer. The Bi2212 molar fraction was estimated from the intermediate peak position of the (002) line in reference of the Bi2212 and Bi2201 pure phase data as described in a previous paper[5].

The electrical resistivity was measured by a conventional four probe method with the applied current of 1

## 3. RESULTS AND DISCUSSION

Figure 1 shows temperature dependence of the sheet resistances in the  $c$ -axis oriented thin films with various compositional ratios of the Bi2212 to Bi2201 phases. The Bi2201 thin film with the stacking of 120 half unit cells exhibits a semiconducting (or insulating) behavior with  $\Delta E_{\text{gap}} = 37$  meV as plotted in Fig. 1.  $T_c$  (onset) of the superconducting samples remain almost constant at 82 K notwithstanding the Bi2212 molar fractional range ( $> 48$  %), while the resistance behavior below  $T_c$  (onset) depends on the molar fraction or the normal resistance. Namely, in 92 mol% thin film  $T_c$  (zero) exhibits approximately 40 K, and in 60-80 mol% it reduces to around 10 K. The normal sheet resistance of this sample attains to 4.1 k $\Omega$ , which is close to 6.5

k $\Omega$ , a quantum universal resistance, and suggests that this 48 mol% mixed crystal would locate just on the border of SI transition.

It is well-known that vortex and anti-vortex pair can be generated by applying transport current to the two dimensional compounds such as BSCCO in the temperature range between the vortex KT transition point,  $T^*$  and the superconducting transition temperature  $T_{c0}$  in terms of the Ginzburg-Landau mean field theory. Since they can move around independently over  $T_{c0}$ , the energy dissipative process is realized, then the resistance never reaches zero although it decreases with the reduction of temperature. On the other hand, below  $T^*$  the vortex is combined with the anti-vortex, and the Lorentz forces with the opposite vector bring attractive interaction with each other, therefore, the resistance vanishes out due to the vortex-anti-vortex pairing. Namely, this exhibits that in the two-dimensional mixed crystal thin films, the temperature where the zero resistance takes place is represented as  $T_{\text{KT}}^V$  based on the KT transition theory. In our BSCCO thin film the temperature was observed at 45 K for the 92 mol% Bi2212 mixed crystal thin film, at 7 K for 82 mol% and at 15 K for 66 mol%. Here, we cannot give an appropriate explanation for the inversion of  $T_{\text{KT}}^V$  between 82 and 66 mol%. A plausible explanation may be given by the cause of the different distribution on the percolation paths among the specimens.  $T_{c0}$  means a phase transition point from the insulating or semiconducting state to the superconducting one, and corresponds to  $T_c$  (onset) in this letter. However, in the case of a granular two-dimensional system the Cooper pair starts to generate by the thermal fluctuations as described in terms of the theories of Aslamazov and Larkin[6] at

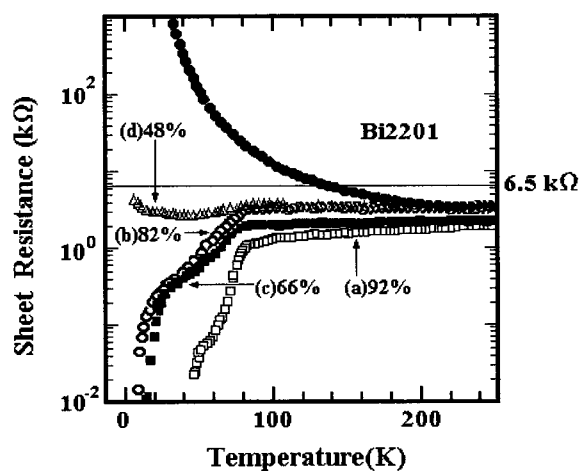


Fig. 1. Temperature dependence of sheet resistance in the (Bi2212/Bi2201) mixed crystal thin films with different Bi2212 fractions.

Thompson[7], therefore,  $T_{c0}$  should be somehow different from  $T_c(\text{onset})$ ,  $T_c(\text{mid})$  where the resistance shows half value at  $T_c(\text{onset})$  is more likely as  $T_{c0}$  according to a proposal of Ota *et al.*[8]. Then,  $T_{c0}$  is given as 74 K for 92 mol%, 62 K for 82 mol% and 63 K for 66 mol%.

In the case where the resistance behavior in the dissipative state is dominated by the vortex KT transition, the temperature dependence of resistance can be expressed as:

$$R = a \exp\left(-2\sqrt{b \frac{T_{c0} - T}{T - T_{KT}^V}}\right) \quad (1)$$

Here,  $a$  and  $b$  are constants. Thereby, the logarithms of sheet resistance  $R_{\square}$  are depicted as a function of  $\{(T_{c0} - T)/(T - T_{KT}^V)\}^{1/2}$  in Fig. 2.

A linear relationship is approximately realized among them for the mixed crystal thin films over 66 mol%. This suggests that the resistance character in these specimens is dominated by the vortex KT transition. Whereas, in the thin film with 48 mol% the superconductive behavior was observed in the temperature range from 50 to 80 K, but below 50 K the semiconducting behavior was replaced, that is, the resistance never reaches zero. This behavior supports an idea that the mixed crystal thin film of 48 mol% Bi2212 locates really on the border of superconducting-insulating(SI) transition as described already from a comparison between the resistance in the normal state

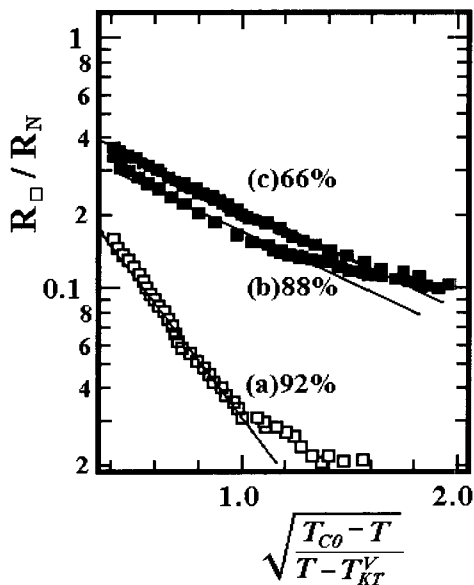


Fig. 2. A linear dependence of the normalized resistance  $R_{\square}/R_N$  on  $\{(T_{c0} - T)/(T - T_{KT}^V)\}^{1/2}$  in the thin films with the different molar ratios.  $R_N$  value corresponds to  $R_{\square} = 1$  ohm at  $T_c(\text{onset})$ .

and the quantum universal.

The resistance upturn below 50 K cannot be ruled out by the vortex KT transition of Eq. (1) in the 48 mol% thin film, but the charge KT transition is considered to be predominantly realized in the lower temperature region. Namely, in this thin film the competitive charge and vortex KT transitions take place corresponding to two different temperature regions. The concave type  $R_{\square} - T$  curve in Fig. 1 is considered due to temperature dependence of the free carrier density,  $n(T)$ , which is represented by the following expression based on the charge KT transition theory[8].

$$n(T) = n_0 \exp\left(\frac{-2b^c}{\sqrt{T/T_{KT}^c} - 1}\right)$$

Here,  $n_0$ ,  $b^c$  are constants, respectively. As the sheet conductance,  $G_{\square}$ , is in proportion to this carrier density  $n(T)$ , it is given by

$$G_{\square} = G_0 \exp\left(\frac{-2b^c}{\sqrt{T/T_{KT}^c} - 1}\right)$$

The semiconducting behavior of the sheet resistance  $R_{\square}(T < 50 \text{ K}) = G_{\square}^{-1}$  for the 48 mol% thin film is well fitted by Eq. (3) using the least mean squares method and shown in Fig. 3. The charge KT transition position  $T_{KT}^c$ , and a constant,  $b^c$ , were calculated as fitting parameters and obtained to be 0.10 K and 1.9, respectively. However, the obtained value of the  $b^c$  seems to be too large since it is generally close to 1. Thus,  $T_{KT}^c$  and

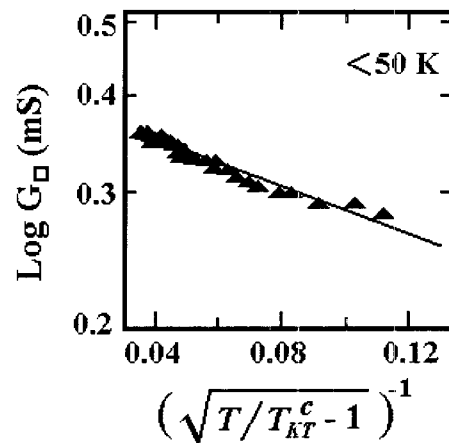


Fig. 3. Temperature changes of the sheet conductance  $G_{\square}$ , in the 48 mol% mixed crystal thin film below 50 K and its least-squares-fitting line by Eq. (2).  $T_{KT}^c = 0.10$  K assuming that  $b^c = 1$ .

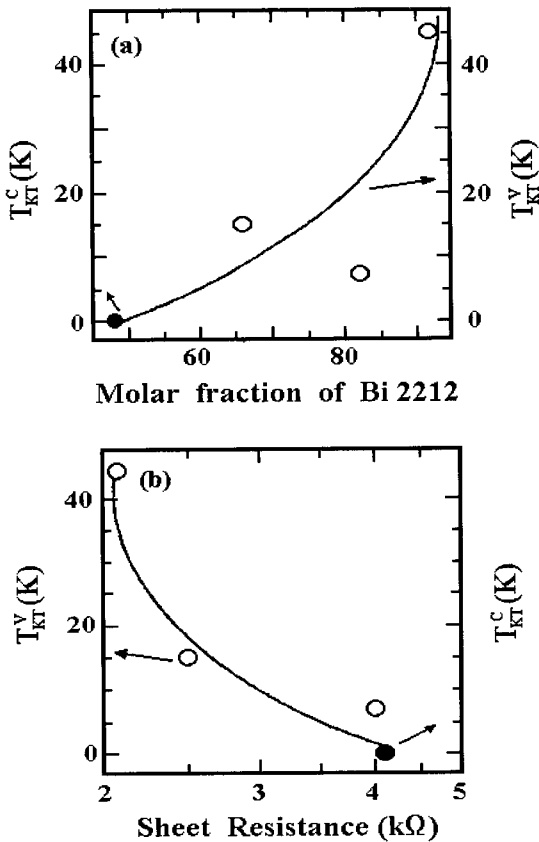


Fig. 4. The correlation between KT transition temperature and (a) molar fraction of Bi2212 phases or (b) sheet resistance.  $\circ$  :  $T_{KT}^v$ ,  $\bullet$  :  $T_{KT}^c$ .

estimated at 0.16 K, assuming that  $b^c = 1$ , and there is little difference of the standard deviation between both calculations. Accordingly,  $T_{KT}^c$  will be about 0.16 K. This fitting means that in the 48 mol% mixed crystal thin film  $R_{\square}$  below 50 K is dominated by the vortex state. Both KT transition temperatures,  $T_{KT}^c$  and  $T_{KT}^v$ , are plotted against molar fraction of the Bi2212 in Fig. 4(a) and against the sheet resistance in the normal state in Fig. 4(b). Fig. 4(a) exhibits that the charge KT transition becomes predominant with the increase of the Bi2212 insulating phase and that the discrimination of transition takes place at 48 mol%.

#### 4. CONCLUSION

It was found that the boundary of SI transition coincides at the 48 mol% in the BSCCO mixed crystal thin films. Moreover, it was clarified from the sheet resistances that the vortex and charge KT transitions could take place competitively on the process of temperature variation even in a specimen with 48 mol%. The latter is depressed as temperature increases,

while the former predominantly emerges. The critical point  $T_{KT}^c$  was realized at very low temperature because of this competition. It turns out that in Fig. 4(b), the vortex KT transition temperature reduces as the sheet resistance  $R_{\square}$  enhances and around 4.1 kΩ, the vortex KT transition has exhausted.

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