Normal-state reentrant behavior in superconducting $Bi_2Sr_2CaCu_2O_8/Bi_2Sr_2Ca_2Cu_3O_{10}$ intergrowth single crystals

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Normal-state reentrant behavior is observed in the resistive transition for Bi-2212/2223 intergrowth single crystals. It is found that for a small driving current of 50 μA the resistivity for the intergrowth system exhibits a superconducting transition with zero values at 105 K, which returns to the resistive state at a lower temperature. With a further temperature decrease, the system again becomes superconducting at 92 K. In the case of large driving currents, the reentrant behavior is not found and only a two-step resistive transition is observed. The normal-state reentrant behavior is interpreted using a model where the Josephson coupling between the CuO_2 trilayers is weakened by thermally activated resistance between these layers. The two-step transition can be explained by the proximity effect along the *ab* plane. Our results suggest that due to thermally activated resistance between the superconducting layers in the layered superconductors like Bi-Sr-Ca-Cu-O, the three- to two-dimensional crossover of superconducting behavior may be induced at temperatures much lower than T_c by applying magnetic or electric fields.

INTRODUCTION

A characteristic structural feature of the high-T_c superconducting cuprates is the presence of CuO_2 planes which strongly suggests layered or two-dimensional physical properties. Among these cuprates, $Bi_2Sr_2CaCu_2O_8$ (BSCCO) was found to show highly anisotropic behavior in the superconducting transition,[1] normal-state resistivity,[2] thermal fluctuation,[3] upper critical field,[4] and transport critical currents.[5] Recently, some unusual transport phenomena which reveal the vortex motion as well as the interlayer Josephson coupling of the thermally excited vortices of this layered structure have been reported,[6–8] and a direct measurement of the ac and dc Josephson effects between CuO_2 bilayers has been made with the current flow along the *c* axis.[9]

As is well known, the normal-state resistivity along the *c* direction, ρ_c, of BSCCO is several orders of magnitude larger than that along the *ab* plane, ρ_{ab}, with $\rho_{ab}(T)$ being typically metallic while $\rho_c(T)$ has a thermally activated temperature dependence similar to that of a semiconductor. This semiconductor-like behavior can extend to temperatures much lower than its mean-field superconducting critical temperature T_c as the superconductivity of the system is suppressed by applying a strong magnetic field.[10] Therefore, the BSCCO system can be regarded as a layered system in which the superconducting CuO_2 bilayers are separated by semiconductor layers. Since the system is quasi-two-dimensional along the *ab* plane, the in-plane thermal fluctuation should be very strong. As observed by Wan *et al.*,[7] the development of a bulk zero-resistance state takes place through a sequential process: with decreasing temperature, the Josephson interaction couples the two-dimensional CuO_2 bilayers at a temperature T_c^c, then, at a lower temperature designated as T_c^{ab}, the bilayers undergo a Kosterliz-Thouless-type transition to a zero dissipation state. However, considering the thermally activated temperature dependence of $\rho_c(T)$, the Josephson interaction between the CuO_2 bilayers is weakened with decreasing temperature, and thus it is easily destroyed by applying an external field, leading to a three- to two-dimensional crossover of superconducting behavior. This type of crossover has been observed by Nakamura *et al.*[11] In principle, the resistive state along the *c* axis should appear if the Josephson coupling between the bilayers is fully destroyed. Here, we address this issue through the measurement of the superconducting transition of $Bi_2Sr_2CaCu_2O_8/Bi_2Sr_2Ca_2Cu_3O_{10}$ intergrowth single crystals. As described below, these Bi-2212/2223 intergrowth single crystals provide us with a special system of layered high-T_c superconductors in which superconducting CuO_2 trilayers are spaced apart a distance ranging from 1 to 40 unit cells of Bi-2212. Since the distance between the trilayers in this intergrowth system is wider than that between CuO_2 bilayers in the pure Bi-2212 system, the Josephson coupling between the superconducting layers in this intergrowth system is weaker than that in the pure Bi-2212 system, and thus it is more easily destroyed. On the other hand, the CuO_2 trilayer system has a $T_c \sim 110$ K, roughly 20 K higher than that of the CuO_2 bilayer system, and thus it is possible to investigate the decoupling behavior of the superconducting trilayers before the Bi-2212 matrix becomes superconducting. It is found that the intergrowth system shows a superconducting transition (zero resistance) at 105 K, which then returns to a resistive state at a lower temperature due to decoupling between the superconducting layers. This reentrant phenomenon systematically varies with the driving current and applied field.

EXPERIMENTAL

The $Bi_2Sr_2CaCu_2O_8/Bi_2Sr_2Ca_2Cu_3O_{10}$ intergrowth single crystals measured in the present experiments were prepared by a traveling solvent floating zone method. The growth details have been described elsewhere.[12] Intergrowth single crystals were selected from an as-grown rod of $Bi_{2.1}Sr_{1.9}Ca_1Cu_2O_x$. In order to guarantee that the chosen crystals are single crystals, free of any low-angle grain boundaries, they were extracted from polished cross sections of the rod under a polarizing microscope. Laue x-ray diffractometry revealed that the samples were good single crystals. However, it is interesting to note that high-resolution transmission electron microscopy (HRTEM) images revealed some CuO_2 trilayers existing in the Bi-2212 matrix [see Fig. 1(a)]. The HRTEM results show that the CuO_2 trilayers always appear accompanied by an antiphase boundary (APB). The reason for this is that the Bi-2223 layer cannot extend from one side of the crystal to another in only one layer. As soon as the CuO_2 trilayer ends inside the crystal, there will be an APB as shown in Fig. 1(b). The CuO_2 trilayers are certainly in the c plane of the Bi-2212 matrix, and connect with an APB, forming a "defect line." The distance between the trilayers ranges from 1 to 40 unit cells of Bi-2212. Most of the trilayers have a length along the ab plane in the range 100–250 Å. Thus, the crystals can be described as a system in which separated CuO_2 trilayers have been inserted into the Bi-2212 matrix. As the distance between the trilayers is not constant, but randomly distributed between 1 and 40 unit cells of Bi-2212, the trilayers cannot be described as a superlattice of the system. Also, since the length of the trilayers along the ab plane is limited, they are not continuous in the matrix (see the schematic in inset a of Fig. 3). The superconducting transitions were measured by using a SQUID magnetom-

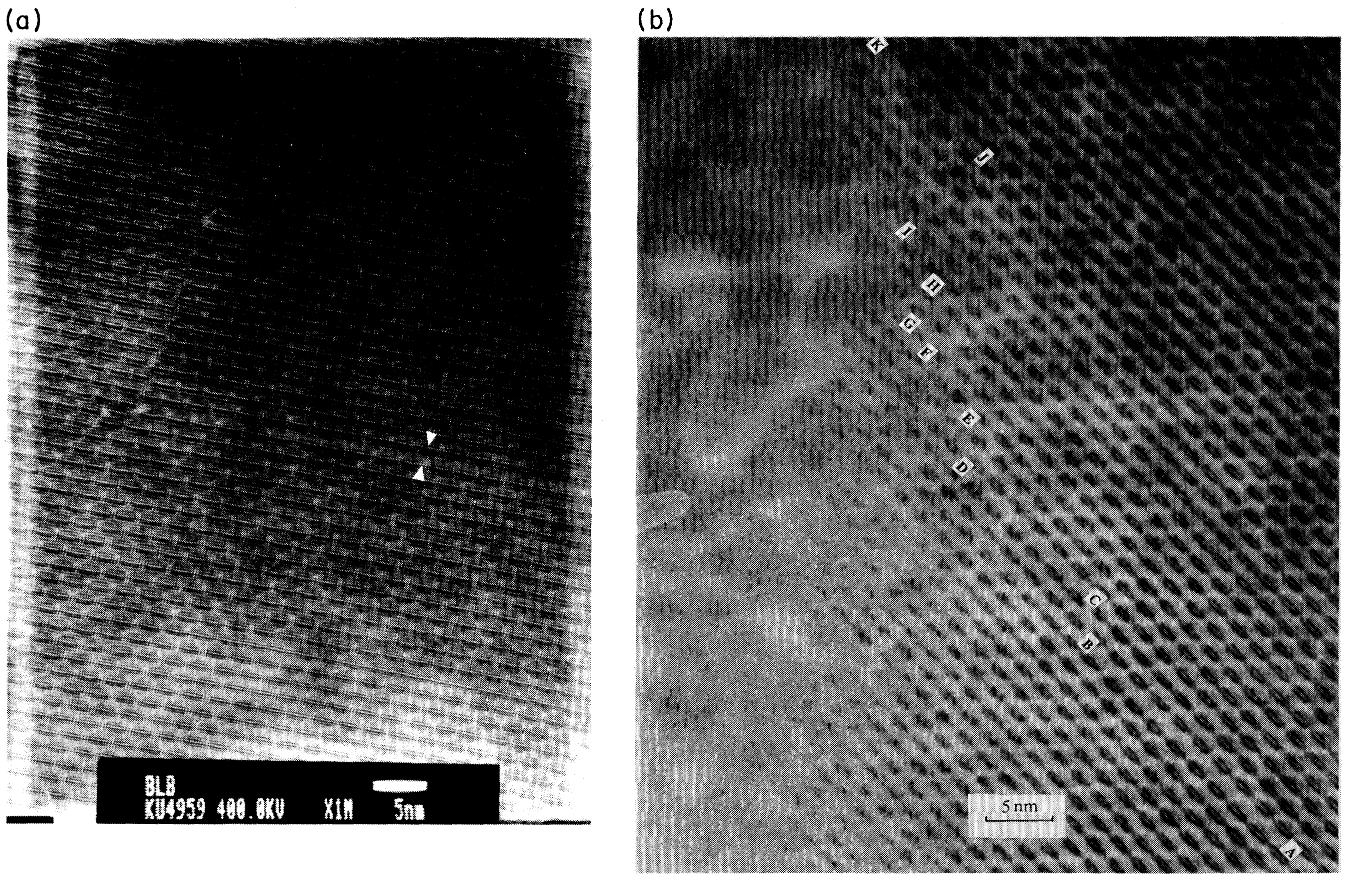

FIG. 1. HRTEM images of the Bi-2212/2223 intergrowth single crystals. (a) The evidence of one single layer of Bi-2223 inside the Bi-2212 matrix. The indicated layer has a width of 18 Å (half unit cell of Bi-2223, other layers have a width of 15 Å (half unit cell of Bi-2212). (b) Typical distribution of the CuO_2 trilayers in the Bi-2212 matrix; shown here are three segments of CuO_2 trilayers labeled as AB, CD, and JK. The lines BC and $DEFGHIJ$ represent the antiphase boundary (APB).

eter. The resistivity measurements were made using both an ac and dc four-terminal technique. The current and voltage contacts were evaporated silver strips on the top surface of the crystal, each having a contact resistance of less than 1Ω.

RESULTS AND DISCUSSION

Figure 2 shows the temperature-dependence magnetization of the sample at a field of 10 Oe as measured by the SQUID magnetometer. A very large Meissner transition appears with an onset at 92 K corresponding to the superconducting transition of the Bi-2212 matrix. The enlarged region of the curve between 90 and 130 K shows a small diamagnetic signal with an onset temperature of ~ 110 K (see the inset of Fig. 1). This signal is assumed to be due to the superconducting transition of the CuO_2 trilayers. This latter signal is typically 1000th as large as that from the Bi-2212 matrix, indicating that the number of CuO_2 trilayers is minimal compared with that of the bilayers of the Bi-2212 matrix, and that the magnetic coupling between the trilayers is very small since the magnetization of each trilayer is zero to first order.

The temperature dependence of the resistivity for a single-crystal sample with the dimensions of $6.50\times3.10\times0.08$ mm^3 (denoted as sample A) is shown in Fig. 3. As pointed out previously for the layered compounds,[6] the current distributes itself inhomogeneously in the sample. The resistivity shown here is merely a nominal one, denoted as R. The sample shows two transition steps, one is at ~ 110 K and the other at ~ 92 K, corresponding to the two transitions observed in the magnetization measurement, which can be associated with superconducting transition of the trilayers and Bi-2212 matrix, respectively. For sake of convenience, these two transition temperatures are denoted as T_{c1} and T_{c2} and their values are taken as 110 and 92 K, respectively, determined from the onset transition temperatures in the magnetization measurements. (The resistive transition curves was not used to determine the values of T_{c1} and T_{c2} as it is sensitive to the driving current.) In the case of a very low driving current (50 μA), the sample shows zero resistance at the first transition which suggests the development of a continuous superconducting path through the trilayer segments in the sample. Upon decreasing the temperature further, the resistive state is returned at 102 K, i.e., a normal-state reentrant behavior appears in the system. However, in the case of a larger driving current or applying a magnetic field the reentrant phenomenon disappears, and the system exhibits only a two-step resistive transition.

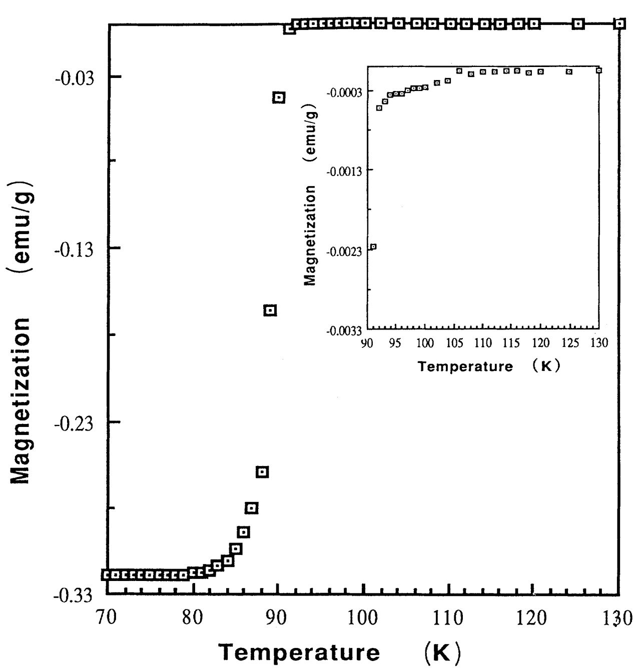

FIG. 2. The temperature-dependence magnetization curve for a typical Bi-2212/2223 intergrowth single crystal. Inset: enlarged region of the curve between 90 and 130 K.

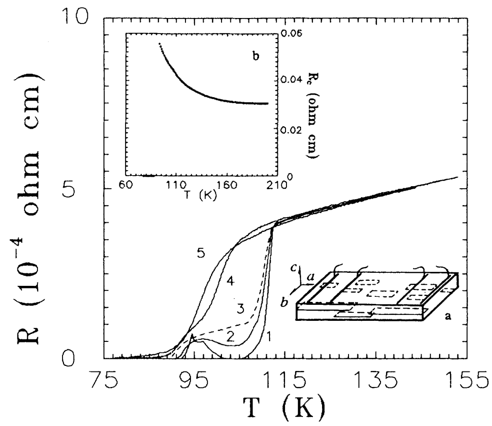

FIG. 3. The resistive transition of a Bi-2212/2223 intergrowth single crystal (sample A) for different driving currents and magnetic fields. (1) 0.05 mA, 0 Oe; (2) 0.15 mA, 0 Oe; (3) 50.0 mA, 0 Oe; (4) 1.0 mA, 5 kOe ($H\|ab$ plane); (5) 1.0 mA, 5 kOe ($H\|c$). Inset a: Schematic of the Bi-2212/2223 intergrowth single crystals. Inset b: Temperature dependence of the resistivity along the c axis, $R_c(T)$.

The normal-state reentrant behavior mentioned above is a common feature of our intergrowth single crystals. Figure 4 shows a similar reentrant behavior for another crystal sample with the dimensions of $4.50\times2.80\times0.08$ mm^3 (denoted as sample B) selected from the same rod as sample A. This sample also shows two transition steps at approximately 110 and 92 K, respectively. Similarly, this sample exhibits zero resistance at 105 K in the case of low driving currents, and the resistive state is returned at 102.8 K with further decreasing temperature. Applying larger driving currents or increasing magnetic fields, the reentrant behavior disappears gradually. It is clear that the main features of the normal-state reentrant behavior for these two samples are the same.

To confirm that the samples actually become superconducting at the first transition and to obtain information concerning the type of transitional behavior that the samples are exhibiting, the I-V characteristic of the samples was measured for temperatures between 100 and 106 K.

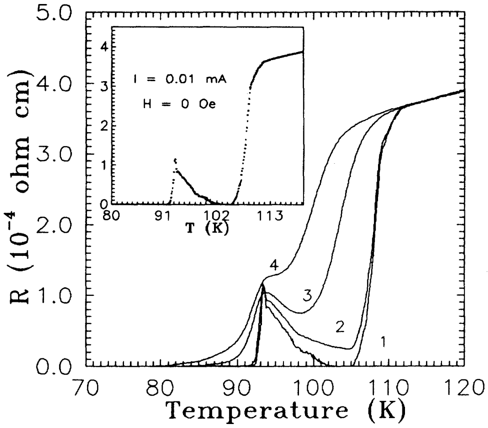

FIG. 4. The resistive transition of a Bi-2212/2223 intergrowth single crystal (sample B) for different driving currents and magnetic fields. (1) 0.01 mA, 0 Oe; (2) 1.0 mA, 0 Oe; (3) 1.0 mA, 200 Oe ($H\|c$); (4) 1.0 mA, 1 kOe ($H\|c$). Inset: Details of the reentrant behavior for condition (1).

Figure 5 shows the I-V curves of sample B at four typical temperatures. All the I-V curves show nonlinear behavior which can be approximately described, in the large current region, by the power law $V \sim I^{\alpha}$ with $\alpha > 1$. There are two further features to note concerning the I-V characteristics shown in Fig. 5. First, the curvature of the ln$E$ vs ln$J$ curve is negative at the temperature where the sample shows zero resistance in the resistive transition (see curve 2 in Fig. 5), but it is positive for the temperature at which the sample is in a nonzero resistance state. The negative curvature is an indication of the true superconducting state whereas the positive curvature indicates the existence of a finite resistance. Second, the parameter α decreases, with decreasing temperature, from 1.85 at 105.5 K to 1.07 at 100.3 K. This means that the sample is close to normal state after the first and before the second superconducting transitions, i.e., between 100 and 94 K.

To understand the mechanism causing the observed normal-state reentrant behavior, we assume, at first, that between T_{c1} and T_{c2}, the CuO_2 trilayer segments are coupled to each other through the proximity effect along the ab-plane direction (since the transport properties in this direction of the Bi-2212 matrix are dominated by the CuO_2 bilayers), resulting in the system being an analogue of a proximity-effect junction array along the ab plane. In addition, according to the results of Busch *et al.*,[6] the current decays exponentially into the crystal and is mainly confined to a thin surface layer of thickness $z_{\rm eff}$, i.e., $j_x(z) \approx \exp(-|z|/z_{\rm eff})$, where $z_{\rm eff} = L\pi^{-1}(\rho_{ab}/\rho_c)^{1/2} \approx 1$ μm in our case (L is the length of the sample). Therefore, the system can be regarded, approximately, as a two-dimensional system. If we denote E_{ab} as the average coupling energy between the CuO_2 trilayer segments along ab plane, then we obtain[13]

$$E_{ab}(T) = \hbar I_c^{ab}/2e$$
$$= E_{ab}(0)(1-T/T_{c1})^2 \exp(-d/\zeta_n) \; , \qquad (1)$$

where d is the thickness of the proximity-effect junction, and ζ_n is the normal-metal coherence length.

The contribution of the proximity effect to the resistive transition is now discussed. As shown in the inset of Fig. 6, the temperature-dependence resistivity curve for the sample shows a drop initially at T_{c1}, followed by a region of gradually decreasing resistivity. A proximity-effect junction would have a nonzero critical current at all temperatures below its superconducting critical temperature but for thermal noise, which acts to destroy phase coherence of the superconducting order parameter in the normal region. This region can maintain weak superconductivity only when the coupling energy E_{ab} is larger than the thermal noise energy $k_B T$. The drop in resistivity is proportional to the fraction of this region to that for the whole system. This gives[14]

$$R(T)/R_0$$
$$= 1 - \beta(\zeta_n/d) \ln[E_{ab}(0)(1-T/T_{c1})^2 \zeta_n / w k_B T] \; , \qquad (2)$$

where R_0 is the resistivity of the Bi-2212 matrix in the ab

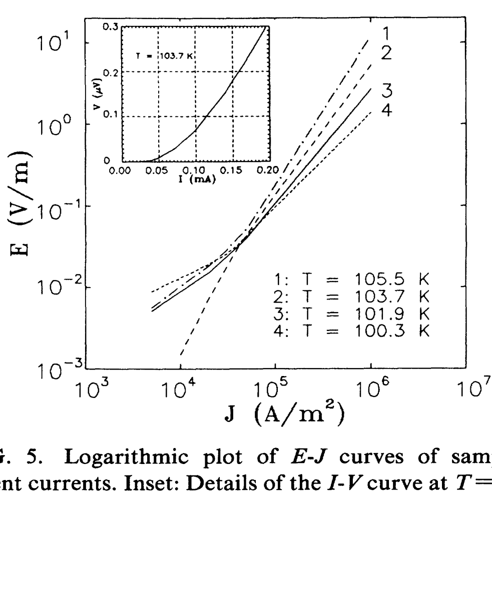

FIG. 5. Logarithmic plot of E-J curves of sample B at different currents. Inset: Details of the I-V curve at $T = 103.7$ K.

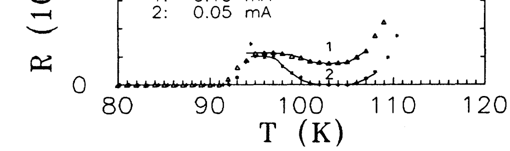

FIG. 6. The reentrant process in the resistive transition at different currents. The solid lines are the fitted results of Eq. (5). Inset: Two steps in the resistive transition. The dashed line represents the fitting of a proximity effect junction array, Eq. (2).

plane before the proximity effect occurs, w the width of the junction, and β is a constant of order unity. Since ζ_n is related to the Fermi velocity v_F and mean free path l by $\zeta_n = (\hbar v_F l / 6\pi k_B T)^{1/2}$,[13] then its value can be estimated to be 30 Å at $T = 100$ K by choosing $v_F = 1.1 \times 10^7$ cm/s and $l = 200$ Å for the typical high-T_c superconductors.[15] The value of w can be estimated from the length of the trilayer segments, and taken as 150 Å. Furthermore, if we assume that the junctions exhibiting the proximity effect are not very thick, then we suppose the thickness of the junction is less than that of the coherence length along the ab plane, i.e., $d = 10$ Å. In this case, $E_{ab}(0)$ and β are the only free parameters. Choosing $\beta = 3$, the fitting between the data and Eq. (2) for sample A is illustrated by the solid line in the inset of Fig. 6 where $E_{ab}(0)$ is fitted as 3×10^{-19} J, a figure equivalent to the critical current $I_{ab}^c(0) = 2eE_{ab}(0)/\hbar = 1$ mA. Using this data we can estimate the gap value $\Delta_{ab}(0)$ as 19 meV,[16] which is close to the 12 meV obtained by Kleiner *et al.*[9] Good agreement between the theoretical curve and the experimental results is seen in the temperature range from T_{c2} to T_{c1}. At a temperature close to T_{c2}, a significant deviation appears. This is due to the fact that for the model proposed above, the Bi-2212 matrix is regarded as a normal metal and its superconductivity is not taken into account. In other words, the model is valid only for the temperature range of T_{c2} to T_{c1}. From the discussion above, it is evident that the proximity effect is the reason for the two transition steps. However, it is not the origin of the normal-state reentrant behavior observed at ~ 103 K. If it was the case, then the continuous superconducting path formed by the proximity-effect junctions would not be destroyed by decreasing temperature, and the zero-resistance state that appeared at 105 K would be maintained at all temperatures below 105 K. Thus, normal-state reentrant behavior would not be observed.

To explain the normal-state reentrant behavior, we believe the Josephson coupling between the CuO_2 trilayers along the c direction must be taken into account. Here we further suppose that the continuous superconducting path formed at 105 K involves both the proximity-effect junction along the ab plane and Josephson junctions along the c direction. It is evident that this model is consistent with the facts that the Bi-2212 matrix is metallic in the ab plane but semiconducting in the c direction before it becomes superconducting. Now we focus on the Josephson coupling between the trilayers along the c direction since the proximity effect along the ab plane has already been discussed in the above paragraphs. As shown by the HRTEM results, the distance between the trilayers along the c direction ranges from 1 to 40 unit cells of Bi-2212. Because of the short coherence length of the high-T_c cuprates, the Josephson coupling most likely takes place between the CuO_2 trilayers with a distance close to or shorter than the coherence length. If we denote E_c as the average interlayer coupling energy between the adjacent CuO_2 trilayers, E_c can be written as[17]

$$E_c(T) = \hbar I_c^c / 2e = (\hbar\pi/4e^2)[\Delta_c(T)/R_n]\tanh[\Delta_c(T)/2k_B T] , \quad (3)$$

where R_n is the junction resistance in the normal state and $\Delta_c(T)$ is the gap parameter at the boundaries of the junction in the c direction. According to the analysis of Deutscher and Müller,[18] for high-T_c cuprates which have a very short coherence length, $\Delta(T)$ should be given by $\Delta_c(T) = \Delta_{c0}(T)\tanh[b/2^{1/2}\zeta(T)]$, where $\zeta(T)$ is the Ginzburg-Landau coherence length, b the gap suppression range, and $\Delta_{c0}(T)$ the bulk gap parameter far from the junction (here it is chosen as the gap parameter of the CuO_2 trilayers). In the present case, the junction resistance in the normal state, R_n, has the same behavior as ρ_c (see inset b in Fig. 3), i.e., $R_n = R_{n0}\exp(-2E_g/k_B T)$. By fitting the data shown in inset b of Fig. 3, we obtained E_g as 0.01 eV. Furthermore, as T approaches T_{c1}, considering $\zeta(T) = \zeta(0)(1 - T/T_{c1})^{-1/2}$ and $\Delta_{c0}(T) \sim \Delta_{c0}(0)(1 - T/T_{c1})^{1/2}$, then Eq. (3) may be written

$$E_c(T) = E_{c0}(1 - T/T_{c1})^2 \exp(-2E_g/k_B T) \quad (\text{for } T \text{ close to } T_{c1}) , \quad (4)$$

where $E_{c0} = (\hbar\pi/16e^2)[\Delta_{c0}(0)^2 b^2/\zeta(0)^2 R_{n0} k_B T_{c1}]$. It should be noted that Eq. (4) is suitable for temperatures between T_{c2} and T_{c1} as the Bi-2212 is merely considered as a nonsuperconducting matrix. As expressed by Eq. (4), the Josephson coupling takes place near T_{c1}. If the coupling is strong enough, and the driving current is lower than the critical current

$$I_c^c(T) = 2eE_c(T)/\hbar = I_{c0}^c(1 - T/T_{c1})^2 \exp(-2E_g/k_B T) ,$$

where $I_{c0}^c = 2eE_{c0}/\hbar$, then long-range superconducting order in the sample can be formed through c-direction coupling of the superconducting segments (trilayers) (see the schematic in Fig. 7), and zero resistance can be developed. With further decreasing temperature, the two factors in Eq. (4) compete with each other, the thermal activation term having a greater effect on the system. As a result, the critical current $I_c^c(T)$ becomes lower than the driving current, consequently forcing the Josephson junction into the resistive state. Thus, as the resistive state is restored along the c direction, superconductivity along the ab plane still exists, confirming the two-

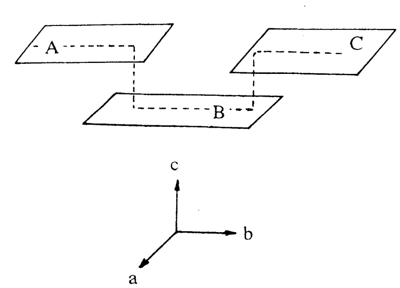

FIG. 7. Schematic of a superconducting path forming through c-direction coupling of the trilayer segments. Here only three pieces of trilayer are shown among which segment A coupled with B along the c direction, then B coupling with C along the c direction, and thus a continuous superconducting path formed from A to C.

dimensional character of superconductivity along the ab plane. According to our model, the resistivity in the reentrant process is proportional to the difference between the driving current and the critical current, i.e., $I-I_c^c(T)$. By further considering the relationship between the voltage in the top surface of the BSCCO crystal sample and the effective driving current,[6] the nominal resistivity in the reentrant process is given by

$$R=\theta[I-I_c^c(T)](\pi D/L)(\rho_c\rho_{ab})^{1/2}[1-(I_{c0}^c/I)(1-T/T_{c1})^2\exp(-2E_g/k_BT)] , \tag{5}$$

where $\theta(x)$ is a step function, and D and L are the thickness and length of the single-crystal sample, respectively. ρ_{ab} and ρ_c are the actual resistivity along the ab plane and c axis, respectively, as determined in Ref. 6. In Eq. (5), I_{c0}^c is the only fitting parameter and the solid curves shown in Fig. 6 are obtained for the indicated driving currents for sample A in which I_{c0}^c was taken as 81 μA. This value is smaller than that for $I_{ab}^c(0)$, but is physically reasonable because the resistivity of the Bi-2212 matrix along the c direction is much larger than that along the ab plane, and thus the coupling between the trilayers along the c direction is weaker. It is evidence that there is good agreement between the theoretical model and the experimental data. Since it is a common feature for the high-T_c cuprates like BSCCO that the superconducting layers are separated by thermally activated resistance layers, our results suggest that due to these thermally activated resistance layers, a three- to two-dimensional crossover of superconducting behavior may be induced at temperatures much lower than T_c by applying magnetic or electric fields.

The influence of a magnetic field can be also interpreted with this model. The only modification is to replace I_{c0}^c with $I_{c0}^c(H)$. The influence of the field is to weaken the Josephson coupling or reduce $I_{c0}^c(H)$, qualitatively, it brings about the same influence on the reentrant behavior as does the driving current.

In summary, normal-state reentrant behavior in the resistive transition for Bi-2212/2223 intergrowth single crystals has been observed. This behavior can be interpreted using a model where the Josephson coupling between the CuO_2 trilayers is weakened by thermally activated resistance between the trilayers. The two-step transition under large driving currents can be explained by the proximity effect along the ab plane.

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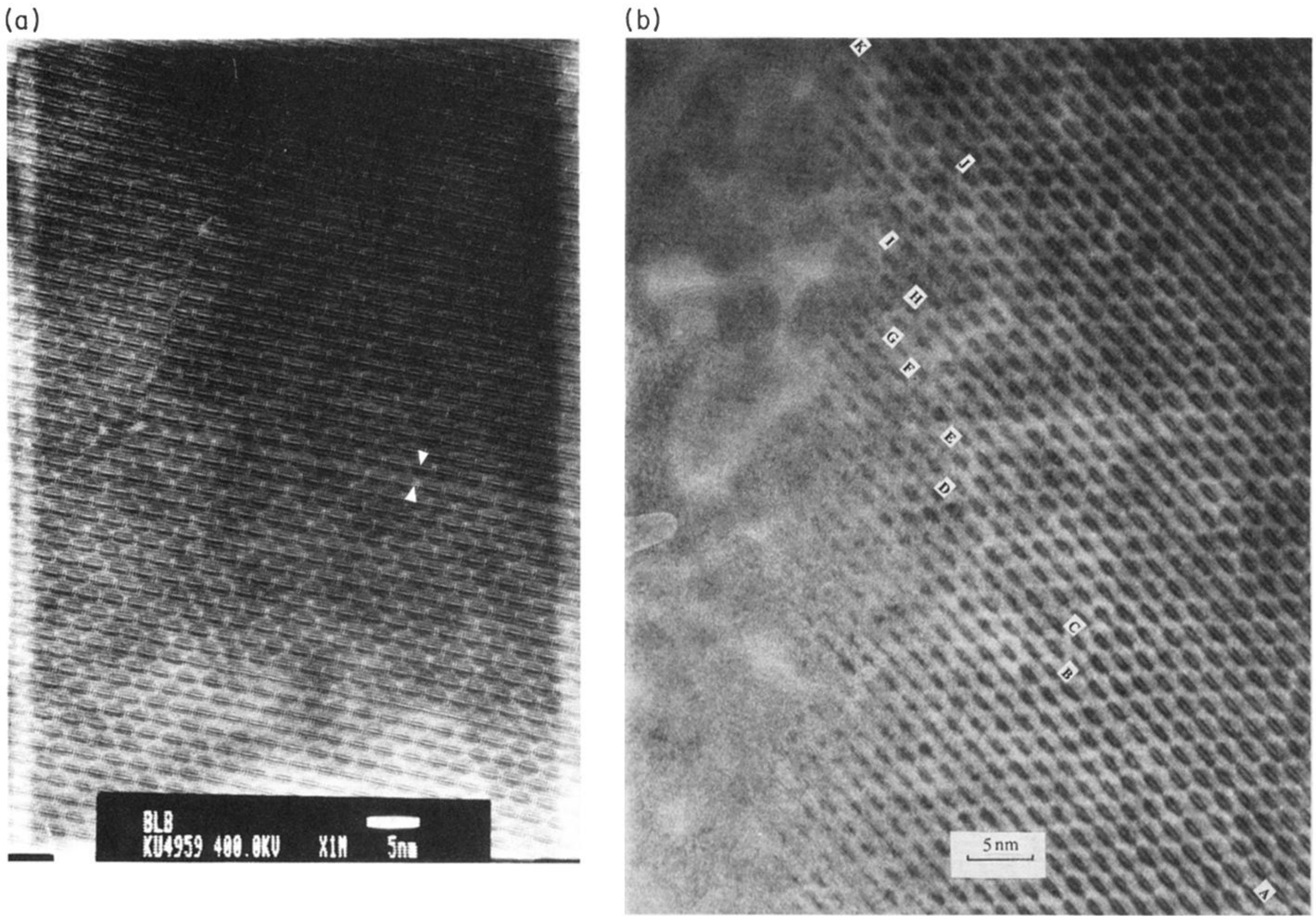

FIG. 1. HRTEM images of the Bi-2212/2223 intergrowth single crystals. (a) The evidence of one single layer of Bi-2223 inside the Bi-2212 matrix. The indicated layer has a width of 18 Å (half unit cell of Bi-2223, other layers have a width of 15 Å (half unit cell of Bi-2212). (b) Typical distribution of the CuO_2 trilayers in the Bi-2212 matrix; shown here are three segments of CuO_2 trilayers labeled as *AB*, *CD*, and *JK*. The lines *BC* and *DEFGHIJ* represent the antiphase boundary (APB).