# $Bi_2Sr_2CaCu_2O_{8+\delta}$ intrinsic Josephson junctions: Surface layer characterization and control

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Intrinsic Josephson junctions on  $Bi_2Sr_2CaCu_2O_{8+\delta}$  superconductors are fabricated using an *in situ* low-temperature cleavage technique, which allows for the fabrication of well-controlled surface CuO<sub>2</sub> double layers of different doping levels. The low-voltage *I-V* characteristics and *c*-axis conductivity properties are discussed for both the surface junctions (*D-I-D'* type) and inner junctions (*D-I-D* type) where *D* denotes a *d*-wave superconductor. Our results show that the experimental data can be satisfactorily described in terms of the theory taking account of the coherent process with a directional tunneling matrix element.

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## I. INTRODUCTION

Intrinsic Josephson junctions (IJJs) are junctions formed naturally within the anisotropic layered high- $T_c$  superconductors, with the CuO<sub>2</sub> layers as their superconducting electrodes.<sup>1,2</sup> Being intrinsic, they have the advantage of avoiding the surface deterioration problem which is often encountered in the other tunneling experiments. In recent years, IJJs have been used widely in the device applications<sup>2</sup> as well as the tunneling spectroscopy studies of the high- $T_c$  materials.<sup>3–9</sup>

In most of the studies, mesa-structured IJJs made on single crystals of  $Bi_2Sr_2CaCu_2O_{8+\delta}$  (Bi-2212) have been used. In this case, all the inner junctions, which are formed between the inner  $CuO_2$  double layers, are identical while the surface junction is different due to the contact of one of its electrodes, the surface  $CuO_2$  double layer, with a metal film. A number of studies have been reported on the inner junctions, which directly reveal the bulk properties of the materials.<sup>3-10</sup> On the other hand, some efforts have been devoted to the understanding of the surface junction properties.<sup>11–16</sup>

The surface junctions can be used as a good probe in the studies of the surface layer properties, as first pointed out by Yurgens *et al.*<sup>11</sup> Kim *et al.* have found suppressed superconductivity in the surface CuO<sub>2</sub> double layers.<sup>12</sup> Later, microwave-induced Shapiro steps in the surface junctions were observed.<sup>13</sup> In these earlier studies, however, the superconducting transition temperature  $T'_c$  of the surface CuO<sub>2</sub> double layer ( $T_c$  will be used to denote the bulk value in this work) is found to be experimentally uncontrollable, which is apparently caused by the surface deterioration during the sample fabrication process.

Recently, we have used a fabrication process in which the Bi-2212 crystals were cleaved *in situ* at liquid nitrogen temperature followed immediately by the evaporation of Au films.<sup>15,16</sup> The low-temperature cleavage was found to give rise to a negligible oxygen loss on the sample surface, which results in an ideal Bi-2212/Au interface. In this paper, we

present a further systematic study utilizing the fabrication process. The surface  $CuO_2$  double layers as well as the  $CuO_2/Au$  interface will be characterized. We will show that the properties of the surface  $CuO_2$  double layers are experimentally controllable, and their doping levels can be adjusted by the fabrication parameters.

Since the fabrication of the mesa containing only a single IJJ is technically difficult, full-range *I-V* characteristics for the surface junctions are not obtainable presently. In this work, the low-voltage *I-V* characteristics and *c*-axis conductivity data, which can be obtained precisely from the branches of the *I-V* curves,<sup>17</sup> will be discussed and compared to the current theoretical treatments for both the inner junctions (*D-I-D* type) and surface junctions (*D-I-D'* type), where *D* denotes a *d*-wave superconductor. Our comparison between the theory and experiment shows that the intrinsic tunneling and *c*-axis transport in Bi-2212 are characteristic of the coherent process<sup>10,18</sup> with a *θ*-dependent tunneling matrix element.<sup>19,20</sup>

### **II. EXPERIMENT**

IJJs on four Bi-2212 crystals (from A to D, see Table I), each of which had three mesas, were fabricated. The single crystals, which were all near optimally doped with  $T_c$ =91.5 K (middle point), were grown by the traveling solvent floating zone method.<sup>21</sup> Small pieces of these crystals with a typical size of  $0.6 \times 0.6 \times 0.05 \text{ mm}^3$  were glued onto Si substrates. The substrate holder used in this work was attached to a liquid nitrogen container in the vacuum chamber, which had a background pressure of  $2 \times 10^{-5}$  Pa. The substrate temperature was measured to be  $\sim 80$  K. For the cleavage of the Bi-2212 crystal, we connected a wire from a piece of sticky tape on the crystal to the shutter. In this way, the Au films, with thickness of 50 to 100 nm, could be evaporated immediately to the fresh surface of the crystal upon cleavage. The whole fabrication process for the IJJs,  $5 \times 5 \ \mu m^2$  in sizes, was rather conventional and has been described in detail previously.14-16

TABLE I. Parameters of four samples made of crystals of the same properties ( $T_c=91.5$ K). There are three mesas on each sample crystal with the parameters listed in three different subcolumns. Note that for samples A through D, the Au-film evaporation rate r is progressively slower. Here, N is the number of IJJs in the mesa. See text for the definitions of  $R_s$  and  $R_b$ .

Sample	<i>r</i> (nm/s)	$T_c'(\mathbf{K})$	$R_S(\Omega)$	$R_b(\Omega)$	Ν
А	6.3	78.7/78.5/79.3	4.85/4.69/3.70	5.88/2.16/3.47	5
В	3.3	72.6/72.5/72.8	8.97/7.70/6.79	1.25/2.82/0.72	7
С	0.74	58.9/58.9/59.1	11.4/11.5/10.1	0.72/0.26/0.78	5
D	< 0.3	14.9/15.9/12.3	18.4/20.4/19.4	1.11/0.36/3.83	6

Three-terminal configuration was used for the *I-V* curve measurements in our experiments in which the Au film was used as both the current and voltage leads. As a result of the configuration, a special circuit was designed for the compensation of the resistance  $R_b$  containing both the CuO<sub>2</sub>/Au interface and lead contributions. The *I-V* curves were recorded using a dc method or by an oscilloscope. The measurements for the resistance R(T) across the mesas were performed using a 1- $\mu A$  ac excitation current and lock-in amplifier.

Figure 1 shows the *I*-*V* curve of the first two branches of sample *A* at T=13 K with  $R_b$  compensated. The first branch in the figure is simply the surface junction *I*-*V* curve and the inner junction data can be obtained by subtracting the voltage values of the first branch from those of the second one for each given current. The conductivity data were obtained from the *I*-*V* curves in the limit of  $V \rightarrow 0$ .

#### **III. RESULTS AND DISCUSSION**

### A. Properties of the surface CuO<sub>2</sub> double layers

Samples A to D listed in Table I were prepared with the same fabrication conditions except that the evaporation rate r of the Au films was different. In Fig. 2, we show the R(T) data for the samples with  $R_b$  subtracted (only the data of one mesa for each sample are plotted for clarity). With decreasing temperature, R is first seen to increase and undergo a fast drop at bulk  $T_c \sim 91.5$  K. The resistance  $R_s$  at the end of the drop represents solely the value from the surface junction. R after that increases again with decreasing temperature until  $T'_c$  below which the surface CuO<sub>2</sub> double layers become superconducting. For the four samples with different r, we can



FIG. 1. Oscilloscope traces of the *I*-V curve of sample A showing the first two branches at T=13 K.

see a clear dependence of  $T'_c$ , which is plotted in Fig. 3 for all three mesas upon each given sample. It can be seen that  $T'_c$  increases monotonically with the increase of r and the scattering of  $T'_c$  for each sample is small. In general, a good reproducibility on different sample crystals has been found using the present fabrication process in our experiment.

There are several indications that suggest a more underdoped surface layer for lower r, in addition to the r dependence of  $T'_c$ . First, by comparing  $T'_c$  for two samples with the same *r* but one with an immediate evaporation of the Au film after the Bi-2212 crystal cleavage while the other with a time delay, it is identified that the involvement of the residual gases in the vacuum chamber at the Bi-2212/Au interface is the major cause for the  $T'_c$  reduction.<sup>16</sup> Other causes have smaller influences since with high r we are able to achieve a  $T'_c$  slightly above 80 K for our crystals with  $T_c$ =91.5 K. Since the main residual gas molecules in a vacuum chamber are CO, H<sub>2</sub>, and water, they can in principle combine with an oxygen atom to form a new compound, e.g.,  $CO \rightarrow CO_2$ ,  $H_2 \rightarrow H_2O$ , and  $H_2O \rightarrow H_2O_2$ . The involvement of the residual gases at the Bi-2212/Au interface is, therefore, very likely to result in the takeaway of the oxygen atoms and push the surface  $CuO_2$  double layers into an underdoped state.

The second indication comes from the  $T'_c$  dependence of  $R_s$ , which is also shown in Fig. 3. As can be seen in the figure,  $R_s$  increases with the decrease of  $T'_c$ , which is clear



FIG. 2. Temperature dependence of the resistance across mesas of samples A to D. The data of only one mesa for each sample are shown for clarity. The line with symbols is the zero-bias resistance taken from the first branch *I-V* curves of sample A. Note that the extension of the line above  $T'_c$  would have basically the same value as that from the direct resistance measurement.



FIG. 3. Superconducting transition temperature of the surface  $CuO_2$  double layer versus the evaporation rate of Au films and mesa resistance taken at the temperature right below  $T_c$ . Lines are guides to the eye.

evidence that the  $CuO_2$  double layers become progressively underdoped as the rate *r* gets slower.<sup>22–24</sup>

It is well known that the superconducting energy gap  $\Delta$  inferred from the sharp peaks in the dI/dV curves increases as the doping level decreases.<sup>25</sup> For sample A, the peak locates at 255 meV. Considering the junction number N=5, we have  $\Delta=25.5$  meV if we disregard the difference between the surface and inner layers for this sample. If we take  $\Delta=25.5$  meV to be the energy gap for the inner layers, we are able to find the values of 36.4 and 57.5 meV for the surface layers of samples C and D, respectively. This shows consistently the same trend as the above data although for our 5- $\mu$ m-sized IJJs, the self-heating effect can be severe near the gap voltage and the data should not be considered to be accurate.



FIG. 4. Resistance versus temperature across the metal films and surface layers of Bi-2212 crystal, which is used to check the interface property. The upper inset shows the sample geometry. The lower inset is the I-V curve at 4.2 K.

# B. The CuO<sub>2</sub>/Au interface

In order to study the  $CuO_2/Au$  interface properties, we carried out a measurement similar to that used by Ekin *et al.*<sup>26</sup> Figure 4 shows the results for a sample prepared with a fast r. The upper inset in the figure shows the sample geometry. On top of the Au-film surface of a Bi-2212/Au structure prepared in the same way as that for the IJJs, we made a SiO<sub>2</sub> window with a width of 8  $\mu$ m and a length of 0.5 mm. The length is in the direction perpendicular to the paper. Across its width, several Ag-film strips,  $3 \mu m$  in width, were deposited. In this way, a four-terminal measurement can be made and the resistance across the thickness of the Ag and Au films and the Bi-2212/Au interface can be obtained. In Fig. 4, we can see that with decreasing temperature, it shows two sharp drops at  $T_c$  and  $T'_c$ . Below  $T'_c$ , the resistance reaches a value of  $\sim 30 \text{ m}\Omega$ , corresponding to a specific resistivity  $\rho \sim 1.5 \times 10^{-8} \Omega$  cm<sup>2</sup> if an effective area of twice the area of  $3 \times 8 \ \mu m^2$  is considered (see Ref. 26). This value sets an upper limit of the interface resistance, which is comparable to the values from the other report<sup>11</sup> and to the result obtained on other materials.<sup>26</sup> The lower inset in Fig. 4 shows the *I-V* characteristic at 4.2 K in which a linear dependence of current upon voltage is seen up to  $\sim 3$  mA. This current magnitude is still considerably larger than the scale of the I-V curve branches in Fig. 1 after taking account of the effective area. Therefore, although it is not known whether the Au film close to the Bi-2212 surface is by the proximity effect superconducting or not, it is clear that the interface resistance is at least small and is a constant well above the current-voltage range of the *I-V* curve branches.

In Fig. 2, the resistance at  $V \rightarrow 0$  taken from the firstbranch (surface junction) *I-V* curves of sample *A* is plotted (line with symbols). It can be seen that the data approach the R(T) curve without any abrupt change as the temperature increases up to  $T'_c$ . This result indicates that as the surface CuO<sub>2</sub> double layer switches from the superconducting to the normal state, there is no significant change of the interface resistance and it remains to be a small value compared to that of the surface junction. The same results are also found for other samples. Our conclusion from these results is that the *I-V* characteristics of both the surface and inner IJJs can be safely obtained after subtracting  $R_b$  for all temperatures up to, or slightly above  $T'_c$ .

## C. Low-voltage I-V curves and c-axis transport

In the studies of intrinsic tunneling and *c*-axis transport properties, there are two aspects that are of much interest. One is whether the process is coherent or incoherent.<sup>10,18,22</sup> The other is the angle dependence of the tunneling matrix element.<sup>19,20</sup> Below, we briefly discuss our data in the light of the current theoretical understandings.

Latyshev *et al.*<sup>10</sup> have investigated the *c*-axis transport properties in IJJs and have found a significant contribution from coherent tunneling. In their treatment, when only coherent tunneling is considered, the quasiparticle decay rate  $\gamma$ is given by



FIG. 5. Experimental *I-V* curves at several temperatures of an inner junction (top) and the corresponding surface junction (bottom) from sample *A*. The curves of two surface junctions from samples *C* and *D* at 4.2 K are shown as lines with open symbols. The inset shows  $\gamma \sim T$  for the inner junction, obtained from the fit using Eqs. (1) and (2).

$$\gamma = \sqrt{\alpha e^2 / 24\beta},\tag{1}$$

where  $\alpha$  and  $\beta$  are the parameters appearing in the functional

$$I = \alpha V + \beta V^3, \tag{2}$$

which is used to fit the low-voltage *I*-*V* curves. The temperature dependence of *c*-axis conductivity  $\sigma$  is described by

$$\sigma(T) \sim 1 + cT^2 \tag{3}$$

for the temperatures below  $T^{\star} \sim \gamma$ .

Our experimental *I-V* curves for the inner and surface junctions are shown in Fig. 5. From fitting the inner junction data using Eq. (2), we can obtain  $\alpha$  and  $\beta$ , and  $\gamma$  is found to be 1.8, 2.35, 4.1 meV for T=4.2, 32, 41 K, respectively. These values are comparable to  $\gamma \sim 3$  meV obtained by Laty-



FIG. 6. Zero-voltage conductivity versus temperature for the surface junctions of samples A, C, and D (solid lines). The data for the inner junction are shown as a dashed line. In the inset, the data are plotted for samples A and C against  $(T/T_c)^3$ , where  $T_c = 91.5$  K.

shev *et al.* for their particular samples.<sup>10</sup> The temperature dependence of  $\gamma$  is plotted in the inset of Fig. 5.

For sample A,  $T'_c$  is closest to  $T_c$ , so the *I-V* curves of the surface and inner junctions do not show much difference, as can be seen in the bottom and top panels of Fig. 5. For samples C and D, however, the curves of the surface junctions become progressively flatter (bottom panel). This can be the result from a junction resistance increase as well as an increase of the superconducting energy gap  $\Delta$  of the surface layers.

In Fig. 6, we show the experimental data of  $\sigma(T)$  for the inner junction (dashed line) and surface junctions (solid lines) for samples *A*, *C*, and *D*. From the experimental results in the low-temperature regime, we find that

$$\sigma \sim T^{\alpha},\tag{4}$$

where  $\alpha$  is ~4.5 for both *A* and *A* (inner) and is ~3.2 for *C* in the temperature range 4.2–35 K. For sample *D*,  $\alpha$  is found to be ~2.9 in the range 4.2–25 K in which the data are available. From these results, we see a clear tendency of decreasing  $\alpha$  for the surface junctions as one of their electrodes gets more and more underdoped. These results deviate from the prediction of Eq. (3), which is found to describe well the data of Latyshev *et al.*<sup>10</sup>

For the *c*-axis properties, Xiang and Hardy<sup>20</sup> have considered the coherent case with a  $\theta$ -dependent matrix element, applying the cold spot quasiparticle scattering model proposed by Ioffe and Millis.<sup>19</sup> The cold spot model separates the scattering rate into the  $\theta$ -dependent and  $\theta$ -independent parts, namely,  $\Gamma_{\theta} = \Gamma_0 \cos^2 2\theta + \tau^{-1}(T)$ . It is found that in the high-temperature regime below  $T_c$ ,  $\sigma(T)$  has a form

$$\sigma(T) \sim T^3 / \Gamma_0, \quad \text{for } T > T^\star, \tag{5}$$

which is independent of  $\tau$ . This means that the  $T^3$  behavior of  $\sigma$  is rather universal, independent of the impurity scattering. In the low-temperature regime,  $\sigma(T)$  is found to be

$$\sigma(T) \sim \tau T^5$$
, for  $T < T^*$ , (6)

which does not depend on  $\Gamma_0$ . However, when impurity scattering in the materials dominates, the  $T^5$  dependence is found to be replaced by a  $T^2$  law.<sup>20</sup>

These results show that the angle dependence of the matrix element plays an important role in the behavior of  $\sigma(T)$ , making it switch from Eq. (3) to Eq. (6). To compare these results with our experimental data, we first note that the above model is proposed for the system in which the superconducting layers have a common order parameter. For the surface junctions having electrodes with different order parameters, the temperature dependencies in Eqs. (5) and (6), apart from the changes of coefficients, are still expected to hold for temperatures below  $T'_c$  (not close to it where the order parameter quickly vanishes).<sup>27</sup> Taking this into account, we find that the model describes satisfactorily our experimental results (with some data near  $T'_c$  included). As can be seen in the inset of Fig. 6, in the high-temperature regime,  $\sigma$  for all the surface and inner junctions falls into the  $T^3$  dependence, as predicted in Eq. (5). This dependence holds in the temperature range from  $T \sim 32$  K up to  $\sim T_c'$  for each given sample and shows its universal character for different samples. The crossover temperature  $T^{\star} \sim 32$  K agrees with the result of Latyshev et al.<sup>10,20</sup>

In the low-temperature regime, we have the result of Eq. (4), with  $\alpha$  ranging from 4.5 to 2.9. For sample *A*, with the cleanest CuO<sub>2</sub>/Au interface,  $\alpha$  is 4.5 for both the surface and inner junctions. This value is close to the prediction in Eq. (6) apart from a possible unknown factor arising from  $\tau^{20}$  For the surface junctions,  $\alpha$  decreases monotonically from sample *A* to sample *D*. Experimentally, from *A* to *D*, more impurities are involved in the CuO<sub>2</sub>/Au interface, leading to

the surface CuO<sub>2</sub> layers more underdoped. Therefore, as is known in the in-plane resistivity measurements,<sup>24</sup> one of the electrodes that constitutes a surface junction should have an increased quasiparticle scattering rate. This will result in the shift of the  $\sigma(T)$  behavior from the case in Eq. (6) toward a  $T^2$  dependence.

#### **IV. CONCLUSIONS**

Intrinsic Josephson junctions on Bi-2212 crystals with well-characterized surface  $CuO_2$  double layers have been fabricated using an *in situ* low-temperature cleavage technique. As verified by their transition temperature, *c*-axis resistivity, and the superconducting gap values, the surface layers can be made progressively underdoped (from samples A to D in Table I) by adjusting the evaporation parameters of Au films. As a result, the surface junctions are composed of the electrodes with different doping levels.

We have discussed the low-voltage *I*-*V* characteristics and *c*-axis conductivity data for both the surface junctions (*D*-*I*-*D*' type) and inner junctions (*D*-*I*-*D* type). Our experimental results support the view that the intrinsic tunneling and *c*-axis transport in Bi-2212 are a coherent process with a directional tunneling matrix element. The temperature dependence of  $\sigma$  for the surface junctions again points to the surface CuO<sub>2</sub> double layers with decreasing doping level from sample *A* to sample *D*.

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