# Andreev reflection in the *c*-axis transport of $Bi_2Sr_2CaCu_2O_{8+x}$ single crystals near $T_c$

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# 임계온도 근처에서 $\mathrm{Bi_2Sr_2CaCu_2O_{8+x}}$ 단결정의 c축 방향 전도시 일어나는 안드레예브 반사

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#### Abstract

An enhancement of the c-axis differential conductance around the zero-bias voltage near the superconducting transition temperature  $T_c$  has been observed in  $Au/Bi_2Sr_2CaCu_2O_{8+x}$  junctions. We attribute such an enhancement to the Andreev reflection (AR) between the surface Cu-O bilayer with suppressed superconductivity and the next superconducting Cu-O bilayer. The continuous evolution of the differential conductance, from gap like depression to an AR-like peak structure, around the zero-bias voltage points to weakening of the barrier strength of the nonsuperconducting layer between adjacent Cu-O bilayers as temperature approaches  $T_c$  from below. The peak structure disappeared just below the bulk  $T_c$  value of underdoped Bi2212 single crystals, whereas it survived up to  $\sim$ 1 K above  $T_c$  in junctions prepared on slightly overdoped crystals. According to a recently proposed theoretical consideration, a wider temperature range of the AR a bove  $T_c$  is expected in the underdoped regime when phase-incoherent preformed pairs emerge in the pseudogap state. Our result is in contradiction to the preformed pair scenario.

 $\textit{Keywords}: And reev \ reflection, preformed \ pairs, \ pseudogap, \ Bi\ _2Sr_2CaCu_2O_{8+x} \ single \ crystals, \ intrinsic \ junctions$ 

#### I. Introduction

The dominant *d*-wave order parameter (OP) symmetry of the high- $T_c$  superconductors (HTSC's) leads to novel phenomena in the electrical transport properties such as the formation of  $\pi$ -junctions [1] and zero-bias conductance peak (ZBCP) [2,3,4]. The

ZBCP is observed in NId (normal-metal/insulator/d-wave-HTSC) junctions when incident quasiparticles (QP's) experience sign changes of the OP upon reflection at the interface of the junction, where a coherent bound state, called the Andreev bound state (ABS) is formed. In contrast to the conventional Andreev reflection (AR) effect [5,6], where maximum conductance enhancement is obtained for zero barrier strength, a finite scattering barrier at the interface is essential for the observation of the ZBCP

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[4].

On the other hand, the c-axis tunneling characteristics the HTSC's such Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+x</sub> (Bi2212) can be well understood in terms of serial stacking of dId intrinsic (d-wave-HTSC/insulator/d-wave-HTSC) Josephson junctions (IJJ's) [7]. Because of the strong barrier strength of the 'I' layers, however, no AR has been observed to date in an NId junction for the c-axis transport. The occurrence of an ABS in such a junction can be ruled out because, in the case of c-axis tunneling, the signs of the OP experienced by the incident and the reflected QP's are always the same.

In this study, we report observation of the conventional AR in the "surface junction" of Bi2212 single crystals for the c-axis transport at temperatures very close to the bulk transition temperature  $T_c$ . In a previous work [8], we showed that the superconductivity of the surface Cu-O bilayer in a Bi2212 single crystal is suppressed when in contact with a normal-metal electrode. Such suppression leads to the formation of a natural NId junction at the crystal surface, the "surface junction", in the temperature range  $T'_c < T < T_c$ , where  $T'_c$  and  $T_c$ are the transition temperatures of the surface and inner Cu-O bilayers, respectively. In this NId surface junction, 'N' represents the surface Cu-O bilayer with suppressed superconductivity including the normal-metallic Au electrode. The fact that the differential conductance (dI/dV) curves evolve in a continuous manner from a gap-like depression at low temperatures to an AR-like peak structure as the temperature approaches  $T_c$  from below leads to a conclusion that the nonsuperconducting 'I' layer between adjacent Cu-O bilayers becomes transparent with vanishing barrier strength at high enough temperatures close to T<sub>c</sub>. A simple numerical simulation using the d-wave BTK formalism [4] confirms, at least qualitatively, our idea of weakening barrier strength near  $T_c$ .

Recently, it has been proposed by Choi, Bang, and Campbell [9] that, if the pseudogap state above  $T_{\rm c}$  in the HTSC's is described in terms of the phase-incoherent preformed pairs (PIPP) [10], the AR can take place from such pairs. They suggest that experimental observation of the resulting conductance enhancement in an ab-planar

normal-metal/HTSC junction would provide unambiguous confirmation of the PIPP in the pseudogap state. Along with the suggestion of Ref. [9] we utilized the appearance of the AR effect along the c-axis direction near  $T_{\rm c}$  to investigate the existence of the PIPP in the pseudogap state. Our result agreed with the recent observation of the nonexistence of the AR effect above  $T_{\rm c}$  in ab-planar normal-metal/HTSC junctions by Dagan et al. [11] and seems to provide negative perspective to the existence of the PIPP in the pseudogap state.

## II. Experiment

As-grown overdoped Bi2212 single crystals were prepared by the conventional solid-state-reaction method. Underdoped crystals were grown first by traveling solvent floating zone methods and the doping level was reduced by annealing the crystals in a low-concentration ( $\sim$ 0.1 %) N<sub>2</sub> mixed O<sub>2</sub> gas. Details of the mesa fabrication are described elsewhere [8,12]. The temperature dependence of the c-axis resistance  $R_c(T)$  and the differential conductance dI/dV were obtained by standard ac lock-in technique.

## III. Results and Discussion

Fig. 1 displays the typical c-axis tunneling resistance  $R_c(T)$  of a mesa (UD2) fabricated on the surface of an underdoped Bi2212 single crystal and measured in a three-terminal configuration as shown in the right inset. The bulk transition temperature  $T_c$ (≈82.5 K) was defined as the peaking temperature of the  $R_c(T)$  curve. As Kim et al. [8] argued earlier, the superconductivity of the surface Cu-O bilayer in Bi2212 single crystals is strongly suppressed when it is in contact with a normal metal layer. The resulting characteristics from the NId surface junction in Fig. 1 is apparent as the strongly semiconducting part of the  $R_{\rm c}(T)$  curve below  $T_{\rm c}$ . However, no superconducting transition of the surface Cu-O bilayer was observed down to ~4.2 K in contrast to the results in as-grown slightly overdoped crystals [8,12], where the surface bilayers usually show the superconducting transition at  $T'_{c} \approx 30-40$  K. In this underdoped crystal UD2, the

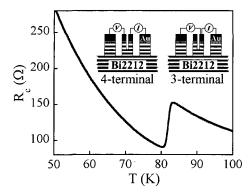


Fig. 1. Typical  $R_c(T)$  curve measured in a three-terminal configuration of the mesa UD2, prepared on an underdoped Bi2212. The maximum  $R_c(T)$  takes place at about 82.5 K ( $\approx T_c$ ). The finite resistance below  $T_c$  was from the surface junction. Insets are schematic views of the sample geometry and the configuration for the three-and four-terminal measurements.

reduced hole doping of the surface Cu-O bilayer [13] may have contributed to the suppression of the superconductivity in addition to the proximity-induced suppression.

A set of dI/dV curves of the mesa UD2 for various temperatures are shown in Fig. 2. The conductance was measured in the first branch of the current-voltage characteristics (IVC), which was from the NId surface junction of the mesa. The dI/dVcurves at the lower temperature range in Fig. 2(a) display distinct gaplike features. Since the IVC of IJJ's in the supercurrent branch switches to the QP branch before the bias voltage reaches the gap edge, sudden jumps appear in the dI/dV curves in Fig. 2(a), where the coupling of the IJJ's broke down before the gap edge of the inner Cu-O bilayer in the NId surface junction was reached. Details of the situation are illustrated in the inset of Fig. 2(b) for  $T \approx 65.2$  K, in which the shaded area approximately marks the region of the voltage jump in the IVC. As the temperature is raised the gaplike depression of the differential conductance becomes shallower [Fig. 2(a)], and gradually exhibits a peak structure [Fig. 2(b)]. However, as shown in Fig. 2(b), in the middle of the superconducting transition slightly above 79.6 K, the magnitude of the peak decreases with increasing temperature and finally vanishes near  $T_c$ . The oscillations of the dI/dV curve with rather

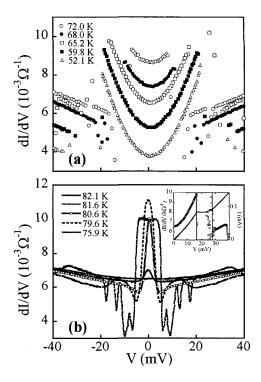


Fig. 2. dI/dV curves of the NId surface junction (a) at relatively low temperatures and (b) at high temperatures near  $T_c$ . Inset shows the IVC (-) and the dI/dV ( $\circ$ ) vs. voltage at  $T{\approx}65.2$  K. The shaded area marks the region of the voltage jump to the QP branch due to the premature switching of the surface junction.

regular periodicity at  $T \approx 75.9$  K are believed to be caused either by the fluctuating Josephson coupling in the IJJ's underneath the surface junction or by the coherent interference of boundary-reflected QP's in the normal-metal electrode of the NId surface junction [14].

The appearance of the zero-bias enhancement of the c-axis dI/dV is quite surprising and in clear contrast to most of earlier results of c-axis tunneling measurements in HTSC's [3,15,16,17], where only gap-like features have been observed. In some exceptions reported, however, the conductance peak was attributed to the possible existence of ab-planar tunneling components due to some surface defects [2,11,18].

The transition from a gaplike structure to a peak in the dI/dV curves was also observed previously in scanning tunneling microscopy (STM) measurements

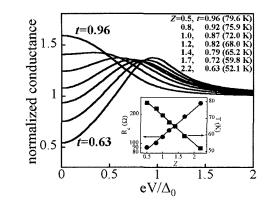


Fig. 3.  $\sigma(E)$  calculated using Equation (3). Fixing  $\sigma(E=0)$  at various temperatures ( $t=T/T_c$ ) to the corresponding measured values, a continuously decreasing Z gives the best qualitative fit to the measured dI/dV curves of Fig. 2. Inset: Temperature dependence of Z ( $\square$ ) and the Z dependence of  $R_c(T)$  (). Z is linearly proportional to  $\log_{10}[R_c(T)]$  and T. Lines are guides to the eye.

on conventional superconductors [19,20]. Reduction the distance between the normal (superconducting) STM tip and superconducting (normal) sample surface turned the conduction characteristics of the system from tunneling-like into weak-link behavior with the tunneling resistance and the corresponding barrier strength Z decreasing. Also, in a c-axis NId junction fabricated by using a sequential deposition of a Bi<sub>2</sub>Sr<sub>2</sub>CuO<sub>6</sub> (Bi2201) thin film and an Au layer on the surface of a Bi2212 single crystal, Matsumoto et al. [21] observed an enhancement of the conductance when the temperature approached  $T_c$ from below. The increase in the conductance was explained due to the conventional AR effect, where the value of the effective barrier strength parameter Zwas reduced with increasing temperature by the semiconducting temperature dependence of the Bi2201 film. We suppose that a similar situation took place in the nonsuperconducting 'I' layer between the surface Cu-O bilayer and the superconducting next bilayer in the NId surface junction. AR between the Au electrode and the surface Cu-O bilayer can be ruled however, since the nonsuperconducting at all the temperatures under consideration.

In order to confirm the appearance of the AR in pure c-axis conduction we followed the formalism by

Tanaka and Kashiwaya [4]. In this case the BTK kernel is given by

$$1 + |A|^2 - |B|^2 = \frac{16(1 + |\Gamma|^2) + 4Z^2(1 - |\Gamma^2|^2)}{|4 + Z^2(1 - \Gamma^2)|^2}, \quad (1)$$

where  $\Gamma = E/\Delta(T,\phi) - \sqrt{[E/\Delta(T,\phi)]^2 - 1}$ . The *d*-wave symmetry of Bi2212 was accounted by  $\Delta(T,\phi) = \Delta(T)\cos(2\phi)$  [22], with  $\phi$  as the azimuthal angle in the Cu-O layer and the temperature dependence of OP magnitude was chosen as [23]:

$$\Delta(T) = \Delta(0) \tanh(3.2\sqrt{T_c/T - 1}). \tag{2}$$

The expression of Eq. (2), which agrees very well with the numerical solution of the BCS gap function except near  $T_c$  where underdoped crystals are known to have a larger gap size [16,17], was obtained by fitting the  $\Delta(T)$  of Ref. [23] to the data of the underdoped Bi2212 crystal with  $T_c$ =83 K in Ref. [17] with  $\Delta(T=0)\approx40$  meV [24]. Using these parameters the normalized conductance was calculated according to the BTK formula [4,6] as

$$\sigma(E) = \frac{4}{4+Z^2} \int_{-\infty}^{\infty} \frac{1}{2\pi} \int_{0}^{2\pi} \left[ 1 + \left| A \right|^2 - \left| B \right|^2 \right] d\phi$$

$$\times \left( -\frac{\partial f(E - eV)}{\partial eV} \right) dE, \tag{3}$$

where f(E) is the Fermi distribution function. The values of Z at different temperatures were chosen by fixing  $\sigma(E=0)$  of Eq. (3) to the measured dI/dV in Figure 2. As shown in the inset of Fig. 3 the values of Z determined in this way are proportional to  $\log_{10}[R_c(T)]$  consistent with earlier observations [20]. In addition, the temperature dependence of Z agrees with the scenario of gradually weakening barrier strength as temperature is increased.

Fig. 3 displays the results of Eq. (3) corresponding to the dI/dV curves of Fig. (2) for T=52.1–79.6 K. The overall qualitative features of the calculated  $\sigma(E)$  have almost one to one correspondence to the experimental data of Fig. 2. At low temperatures, where Z is supposed to be relatively large, the

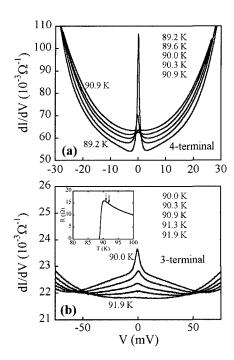


Fig. 4. (a) dI/dV curves of the mesa OD3 measured in the four-terminal configuration and (b) in the three-terminal configuration. Inset:  $R_{\rm c}(T)$  of mesa OD3 obtained in the four-terminal configuration. The two arrows denote the temperatures above which the zero-bias peak of the dI/dV curves in the four- (the left one) and three-terminal (the right one) configurations disappears.

calculated curves show gaplike features that are qualitatively similar to the observed ones. With increasing temperature Z decreases and both the calculated  $\sigma(E)$  and experimental dI/dV curves develop to an AR-like enhancement near the zero-bias voltage in a continuous manner. However, at higher temperatures the measured peak widths are significantly narrower than the calculated ones, which seems to be due to the above-mentioned premature switching to the QP branch of the IJJ's below the NId surface junction. This switching effectively decreases the conductance inside the gap and results in a narrower width of the AR peak. Other possibilities are a decrease in the AR process itself due to thermal excitation of QP's with energies lower than the gap and the thermal smearing of the gap edge very close to  $T_c$ . Both effects decrease the effective OP magnitude experienced by the QP's to a

value smaller than that of Equation (2).

Taking the presence of c-axis AR for granted, we also investigated the c-axis dI/dV curves near  $T_c$  in NId surface junctions made on slightly overdoped Bi2212 single crystals (mesa OD3). The inset of Fig. 4(b) shows the resistive transition of mesa OD3, taken in a four-terminal configuration as illustrated in the left inset of Fig. 1. The temperature of maximum  $R_c(T)$ , which should be close to  $T_c$ , is about 90.3 K and since measurements in a four-terminal configuration do not include the surface effect, the inset of Fig. 4(b) displays the properties of the IJJ's. Fig. 4(a) illustrates the dI/dV curves measured in the four-terminal configuration. The zero-bias peak at each temperature was caused by the Josephson pair tunneling over the stacked IJJ's. With slightly increasing temperature the peak reduces rapidly and disappears completely at 90.9 K, which is defined as the intrinsic value of  $T_c$  of the crystal and marked slightly above the temperature of maximum  $R_c(T)$  by the left arrow in the inset of Fig. 4(b). The concave background conductance implies the existence of the pseudogap in this temperature range [25]. The dI/dV curves measured in a three-terminal configuration from the same central mesa are shown in Figure 4(b). One should note that in this case the properties of the NId surface junction as well as that of the IJJ's were measured. At 90.9 K, where the conductance peak completely vanishes in the corresponding fourterminal measurements, a clear peak structure in the dI/dV curves still persists, although its magnitude is much smaller than the four-terminal counterpart. Increasing temperature the peak height gradually decreases and disappears completely around T=91.9 K as marked by the right arrow in the inset of Fig. 4(b). Thus, in the three-terminal configuration, one has a 1-K temperature window of AR above the  $T_c$ measured by the four-terminal configuration. In a different mesa (OD2) fabricated on another overdoped crystal, a 2-K temperature window of AR above  $T_c$  was obtained (data not shown). Since the inner Cu-O bilayer is no more superconducting above 90.9 K, the AR in Fig. 4(b) is either from the thermally fluctuating superconducting order or from the PIPP in the pseudogap state as proposed in Ref. [9]. If the AR was genuinely from the PIPP, the temperature window of the AR above  $T_c$  should be wider in underdoped crystals than in overdoped ones as pointed out by the authors of Ref. [9]. However, contrary to the theoretical expectation, in the case of underdoped crystals, we observed no appreciable AR above  $T_{\rm c}$  as shown in Fig. 2, whereas a clear temperature window of the AR has been observed in crystals in the overdoped regime.

In summary, we report that an AR has been observed in Au/Bi2212 single crystal junctions with the transport current along the c axis of the Bi2212 HTSC's, in both underdoped and overdoped states. The features of the dI/dV curves changed from gaplike depression at low temperatures to an AR-like enhancement near  $T_c$ . We attribute such continuous evolution to the AR between the proximitysuppressed surface Cu-O bilayer superconducting inner one as the barrier potential decreases with the temperature approaching  $T_c$  from below. We utilized this appearance of the AR to investigate the existence of the PIPP in the pseudogap state above T<sub>c</sub>. In as-grown overdoped mesas we have observed maximally a 2-K temperature range where three-terminal the measurements exhibit the AR-induced conductance peaks above  $T_c$  that was determined in a fourterminal configuration. On the other hand, no appreciable AR above Tc has been detected in single crystals that were in the underdoped regime, although theoretical consideration [9] suggests a wider temperature range of AR above  $T_c$ . Therefore the AR above T<sub>c</sub> in overdoped crystals may have been resulted from the thermal fluctuation effect of the superconducting order. If that is the case no AR due to PIPP in the pseudogap state has been observed in this work. Our results then agree with the recent report in ab-planar junctions [11] where no AR was observed in the pseudogap state. However, more rigorous examination is required on the validity of the suggested observation [9] of the AR effect from PIPP.

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