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Superconducting and Pseudogap States Studied by Using Interlayer Tunneling Spectroscopy on $Bi_2Sr_2CaCu_2O_{8+x}$ Single Crystals

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The existence of a superconducting gap and a pseudogap, and the correlation between them, in cuprates is believed to provide a key to understanding the mechanism of high-T_c superconductivity. In this study, constant-temperature interlayer tunneling spectroscopy measurements were made on stacks of $Bi_2Sr_2CaCu_2O_{8+x}$ intrinsic junctions, excluding the bias-induced self-heating effect. In a certain range of temperatures near T_c, the bias-dependent tunneling spectral distribution showed a peak-dip-hump structure, indicating the coexistence of superconducting and pseudogap states. In a Tesla-range c-axis magnetic field, the spectral weight of the coherence peak diminished, filling mainly the dip region, indicating a major contribution of the dip-region spectral weight to the superconducting condensation.

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I. INTRODUCTION

A pseudogap (PG) behavior, as represented by the depletion of the quasiparticle density of states (DOS), has been revealed in cuprate high-critical-temperature (high-T_c) superconductors in the normal state at temperatures below the characteristic temperature T^* known as the PG temperature [1]. Investigation of this PG behavior and its correlation to the superconducting state are believed to be a key to understanding the mechanism of high-T_c superconductivity. Angle-resolved photoemission spectroscopy (ARPES) and scanning tunneling spectroscopy (STS), both of which are often used powerful surface probes, provide essential information on the quasiparticle DOS as a function of energy with momentum and spatial resolutions, respectively [2]. One of the crucial issues in the recent high-T_c superconductivity research has been to find out whether the existence of the PG is related to the condensation of electronic states to the coherent superconducting state or the PG is a separate ordered state which is irrelevant to the superconducting coherence. A close examination of the temperature and the magnetic-field dependencies of the DOS is expected to provide a clue on these points.

High-T_c cuprates with highly anisotropic layered structures, such as $Bi_2Sr_2CaCu_2O_{8+x}$ (Bi-2212), are known to form a stack of (intrinsic) Josephson tunnel junctions [3]. Interlayer tunneling spectroscopy (ITS) using intrinsic Josephson junctions (IJJs) in Bi-2212 single crystals incorporated with a proper measurement configuration provides the quasiparticle DOS in CuO_2 double layers associated with the junctions located deep enough from the crystal surface. This surface-independent character of ITS measurements is the main advantage of this spectroscopic technique compared to ARPES and STS, which are susceptible to the conditions of the crystal surface. Previous ITS measurements on the temperature dependence of the DOS using Bi-2212 IJJs showed that the superconducting gap closed at T_c while the PG persisted both above and below T_c, indicating the coexistence of the superconducting gap and the PG [4–6]. In a magnetic field, the superconducting gap was observed to vanish close to $H_{c2}(T)$, but the PG remained almost unaltered with H. This field dependence also alludes to the distinct origins of the two gaps [7].

As pointed out recently, however, a serious local heating problem may arise in the ITS, which can easily interfere with the measured spectral distribution [8]. This adverse local heating is caused by the poor thermal conductivity of the Bi-2212 material in the presence of a large bias. Poor thermal conduction between the sub-

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*E-mail: hjlee@postech.ac.kr; Fax: +82-54-279-5564 strate and a stack of Bi-2212 IJJs makes the dissipation of the bias-induced heat even more difficult, enhancing the local heating. This may cast doubts on the credibility of ITS on cuprates. Reduction of the lateral junction size and the number of intrinsic junctions in a stack of IJJs or use of a pulsed bias are the usual schemes adopted to reduce the local heating effect due to the finite bias [4,9]. Any of the above precautions or combination of them, however, turns out to be insufficient to fully exclude the adverse heating effect, which easily causes serious artifact in the ITS measurements.

In this study, we totally eliminated the self-heating effect in ITS by adopting a "heating-compensated" computerized proportional-integral-derivative (PID) feedback control of the sample temperature. Thus, using this scheme, we obtained the genuine electronic spectral distribution without any artifact due to self-heating for stacks of both overdoped and underdoped Bi-2212 IJJs for various temperatures, magnetic fields, and doping levels. We focus on the PG behavior in the normal and the superconducting states, and the possible correlation between them.

The unprecedent high resolution of our ITS allowed us to examine closely the following issues in high-T_c superconductivity. As in the previous ITS measurements, the superconducting gap and the PG are observed to coexist at temperatures close to the superconducting transition temperature T_c at all the doping levels used, as revealed by the peak-dip-hump (PDH) structure in the spectral distribution at temperatures below T_c [4–6]. A sharp coherence peak (CP) is revealed in the superconducting state at its gap edge. In contrast to the predominantly V-shaped DOS previously observed by others [5], the subgap spectrum showed a pronounced U shape, which is consistent with the weighted c-axis tunneling near the antinodal regions of the Fermi surface of the d-wave gap [10]. The high resolution of our ITS, without the interference of local self-heating, indicates that the sharp enhancement of the superconducting CP is mostly contributed by a depletion of the spectral weight of the PG in the bias region just above the superconducting gap edge, which resultantly forms the dip in the PDH structure. This is in clear contrast to the usual BCS case [11], where the spectral weight of the CP is all transferred from the superconducting subgap region. This unusual transfer of the spectral weight may be related to the unconventional electronic character of high-T_c superconductivity.

II. EXPERIMENTS AND DISCUSSION

An as-grown slightly overdoped Bi-2212 single crystal (for the sample OD88) was prepared by using the conventional solid-state-reaction method while the underdoped single crystals were prepared by using the traveling-solvent-floating-zone method (for UD75) [12] and the solid-state-reaction method (for UD77). In this study, a sample was prepared as a 3 × 3 μm^2 stack of IJJs sandwiched between two Au-film electrodes by adopting double-side cleaving of Bi-2212 crystals, micropatterning, and ion-beam etching. The details of the sample fabrication are described elsewhere [13]. The temperature of the sample stack was monitored in-situ by placing another stack of IJJs (the thermometer stack) in proximity to the sample stack, where the two stacks were in strong thermal coupling through the common bottom Au electrode. The lower inset of Fig. 2 shows the sample configuration; the left (right) part represents the sample (thermometer) stack.

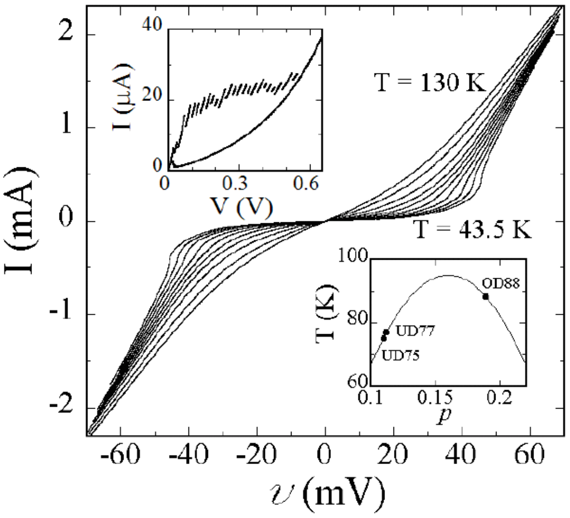

Fig. 1. Tunneling current-voltage characteristics of the underdoped sample UD77 for increasing temperature between 43.5 K and 130 K. The temperatures for the curves from the bottom are 43.5, 49.5, 55.3, 61.3, 67.1, 73, 79.8, 90, 110, and 130 K. Upper inset: Multiple quasiparticle branches obtained at 4.2 K by a single bias sweep. Lower inset: T_c versus the doping level of the samples used.

For constant-temperature ITS, we first raise the sample temperature to a certain value by using a heater coil with a resistance of 230 Ω wound around the substrate holder while maintaining the bath at liquid-helium temperature. The heat generated by the bias current in the sample stack is then dissipated to the thermometer stack through the highly thermal-conductive bottom Au electrode. Monitoring the change in the tunneling resistance of the thermometer stack in comparison with its pre-determined resistive transition data in the low-bias limit allows one to trace the bias-induced temperature variation of the sample stack. Our previous study using heating-compensated ITS confirmed that the sample stack and the thermometer stack in our measurement configuration were, indeed, at an identical temperature with each other [14]. The initial sample temperature is then recovered by reducing the heater current using computerized PID feedback control. The temperature during

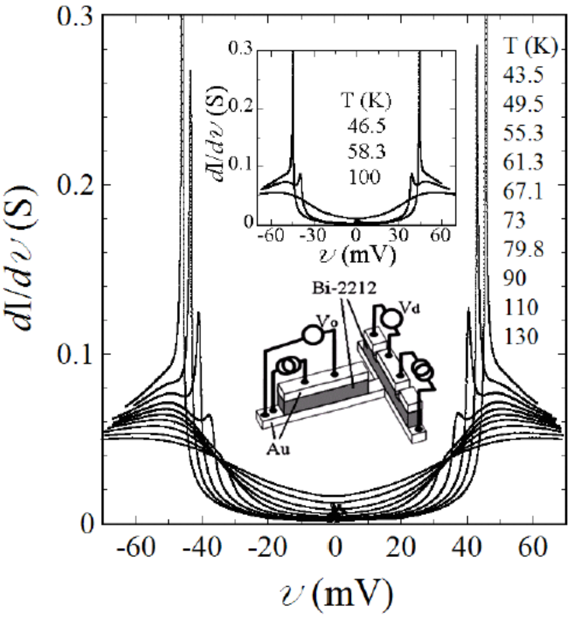

Fig. 2. Interlayer tunneling spectra, dI/dv, for the underdoped sample UD77 as a function of bias voltage per junction at various temperatures corresponding to the curves in Fig. 1 (increasing temperature from top to bottom). Upper inset: Same spectra as in the main panel for three temperatures; far above (100 K), far below (46.5 K), and below (58.3 K) T_c, showing the details of the development of the peak-dip-hump structure. Lower inset: Measurement configuration adopting the in-situ temperature monitor and control scheme.

a sweep was maintained constant within ∼0.1 K in the bias range used in the measurements for all the samples. Measurements were done along the highest-bias quasiparticle curve, *i.e.*, the last quasiparticle branch. The differential-conductance curves were obtained using the conventional lock-in technique at a low frequency of 33 Hz.

The c-axis superconducting transition temperatures, T_c, were 75.2 K (UD75), 77.0 K (UD77), and 88.3 K (OD88), which corresponded to the doping levels of $p = 0.109$, 0.112, and 0.19, respectively (refer to the lower inset of Fig. 1), as determined by the empirical relation, $T_c = 95[1 - 82.6(p - 0.16)^2]$ [6]. The number of quasiparticle branches in the zero-field current-voltage (IV) curves at 4.2 K (see the upper inset of Fig. 1) revealed the total number of IJJs in the sample stacks to be $N =$ 15, 24, and 19 for UD75, UD77, and OD88, respectively.

Fig. 1 illustrates the temperature-dependent gradual variation of the tunneling (IV) characteristics of UD77 from 43.5 K to 150 K. The overall shape of this PID-controlled ITS appears to be similar to the results without heating compensation. The prominent difference between the two cases, however, becomes apparent in the differential conductance. One of the prominent features, even in these IV characteristics, is the absence of the back-bending of the low-temperature curves in the high-bias range, which indirectly, but clearly, indicates that the heating effect is absent from the data.

A series of temperature-dependent interlayer tunneling spectra, $dI/dv(v)$, of the underdoped sample UD77 are displayed in Fig. 2 as a function of bias voltage reduced by the number of junctions as $v = V/N$. As the inset clearly shows, the normal state of the sample above T_c reveals a smoothly depleted DOS in the low-bias range, which is the typical PG feature. For a lower temperature below T_c, a sharp superconducting coherence peak develops inside the PG on the background of the smoothly varying PG feature, constituting the PDH structure. Thus, over a rather wide temperature range below T_c, the SG is seen to coexist with the PG. This is in direct contradiction to the assumption that the presence of the PG state above T_c is a precursor to the emergence of superconductivity below T_c [2]. With further lowering temperature, only the CP becomes apparent as the growing SG size with the fast sharpening CP outweighs the spectrum. In general, the PDH feature becomes more conspicuous for samples with low doping levels. As the CP grows sharply with decreasing temperature below T_c, the position of the dip shifts to higher voltage with deepening depth. Along with this, the low-bias region inside the CP transforms from a V shape to a U shaped. This prominent U-shaped DOS in the subgap region at temperatures below T_c was observed for the first time in this study, which is in clear contrast to the mainly V-shaped DOS observed previously in other studies [5]. The fluctuating conductance at zero bias sufficiently below T_c as seen in Fig. 2 was caused by Josephson pair tunneling.

The U-shaped DOS in the subgap region indirectly suggests that interlayer tunneling is dominated by weighted c-axis hopping of electrons in the antinodal region of the Fermi surfaces of high-T_c Bi-2212 superconductors. The tunneling quasiparticle current in a Josephson junction is expressed as [15]

$$\begin{aligned} I(V) = \frac{1}{2\pi e R_n} \int_0^{2\pi} d\phi |T_\phi|^2 \\ \times \int dE N(\phi, E) N(\phi, E + eV) \\ \times \{f(E) - f(E + eV)\}, \end{aligned} \qquad (1)$$

where R_n is the quasiparticle tunneling resistance, $\phi \equiv \tan^{-1}(k_y/k_x)$, and $N(\phi, E) = \mathrm{Re}\{(E - i\Gamma)/[(E - i\Gamma)^2 - \Delta_0^2 \cos^2 2\phi]^{1/2}\}$ with the quasiparticle scattering rate Γ. It can be numerically shown that the k-independent isotropic tunneling matrix element $T_\phi = 1$ leads to a V-shaped spectral distribution in the subgap bias region [16]. On the other hand, the band calculation for a crystal with a tetragonal structural symmetry, as the Bi-2212 material, predicts that the c-axis quasiparticle tunneling near the antinodal points in the first Brillouin zone is highly weighted so that a tunneling matrix of the form $T_\phi = t_\perp \cos^2 2\phi$ is satisfied [10]. It can be shown numerically that this weighted antinodal hopping reduces low-energy quasiparticle tunneling near the nodal points and leads to the U-shaped tunneling conductance [17]

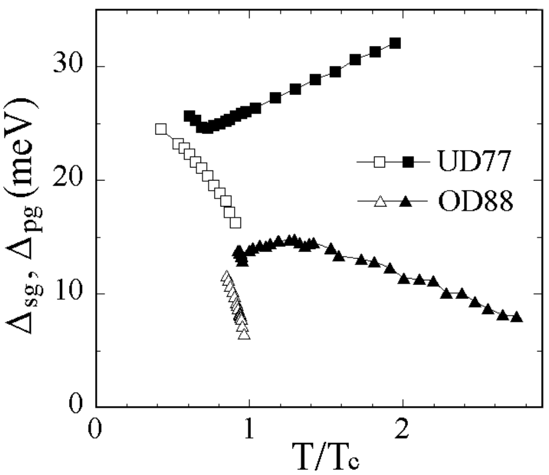

Fig. 3. Temperature dependencies of the superconducting gap (empty symbols) and the pseudogap (filled symbols) for underdoped UD77 (squares) and overdoped OD88 (triangles) samples.

observed in our data. Thus, the pronounced U-shaped spectra in our ITS (Fig. 2) strongly suggest that antinodal hopping is highly weighted in interlayer tunneling. The predominant V-shaped feature observed previously in other ITS studies on low-bias DOS is assumed to be caused by the self-heating effect.

As Fig. 3 shows, for both underdoped (UD77) and overdoped (OD88) samples, the coexistence of the SG and the PG is clearly revealed in the temperature dependence of the SG energy, Δ_{sg} (open symbols), and the PG energy, Δ_{pg} (filled symbols). The superconducting gap closes at T_c, as the superfluid density vanishes. By contrast, the Δ_{pg} exhibits a more complicated temperature dependence. For both samples, Δ_{pg} exhibits a local minimum in the superconducting state slightly below T_c. With increasing temperature, the Δ_{pg} keeps increasing for UD77. For OD88, however, a local maximum of Δ_{pg} is revealed, and Δ_{pg} decreases as the T^* is approached. UD77 is also expected to show similar behavior as OD88, over a much expanded temperature scale. Although it is beyond the scope of this report, the increase in Δ_{pg} above T_c can be qualitatively explained by the PG-related quasiparticle lifetime broadening.

Fig. 4 illustrates the variation of the spectral distribution for the underdoped sample UD75 for increasing magnetic field up to 6 T at two temperatures below [Figs. 4(a) and 4(b)] and at one temperature above T_c [Fig. 4(c)]. For zero field, the sharp CP diminishes rapidly as T_c is approached from below, which is caused by a weakening of the superconducting coherence. A similar suppression of the CP is brought about by applying the perpendicular magnetic field along the c axis. In this sample, the low-temperature curves show much pronounced dip features. One may wonder whether the dip is caused by a simple superposition of the CP and the hump or by a transfer of the spectral weight between them. In the PDH structure, the DOS in the hump drops suddenly at the dip position while sharpening the CP, which becomes more distinct at lower temperatures. Thus, the temperature-dependent spectral redistribution in Fig. 4 strongly suggests that the spectral weight of the CP in the superconducting state is transferred mainly from the dip position in the PG state.

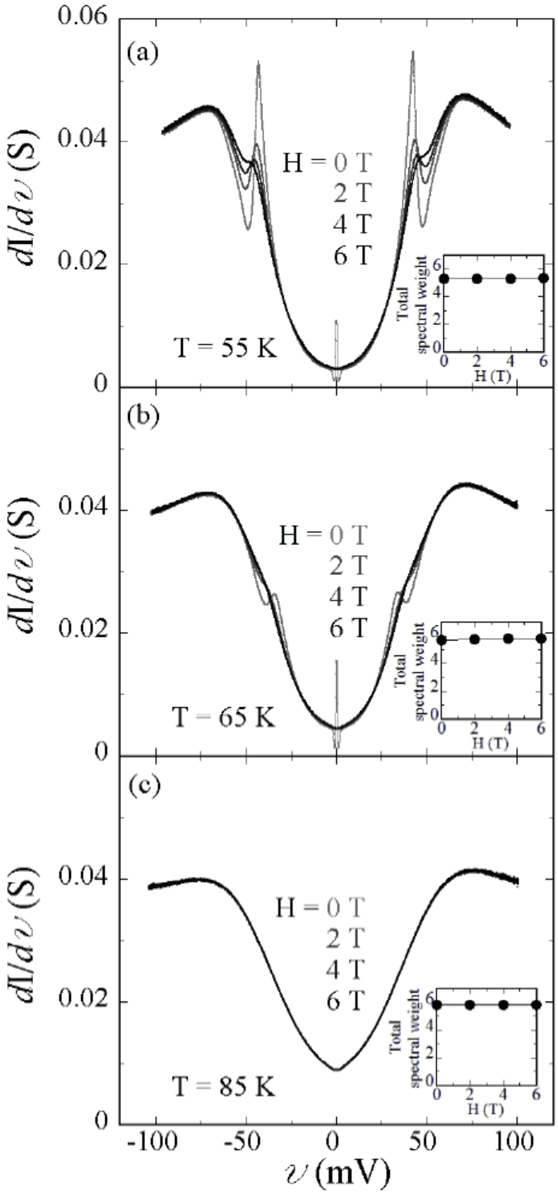

Fig. 4. Field dependencies of the spectral distribution of UD75 at (a) 55 K, (b) 65 K, and (c) 85 K. The inset of each figure shows the total spectral weight for the magnetic fields at the given temperature.

The lower insets of Figs. 4(a), (b), and (c) show the total spectral weight, obtained by integrating the respective spectral-distribution curve for each H field at different temperatures as in the main panel between the two cutoff voltages ($-v_c$, v_c) and normalizing it by the corresponding zero-field value. The cutoff voltage v_c of integration was chosen well beyond the merging position of the different-field spectra around the hump. The resulting total spectral weight turns out to be essentially field independent, indicating that the sum rule holds [18]. This confirms the reliability of our description for the transfer of the spectral weight based on our ITS.

The spectra of UD75 in Fig. 4(a), at a fixed temperature of 55 K, reveal that the CP is suppressed rapidly while the dip fills up with increasing H field. A similar trend holds for the overdoped OD88 with a smeared PDH structure. In comparison, in the process, only a small portion of the CP spectral weight is transferred to the subgap region, especially for UD75. Even at higher temperature, *i.e.*, 65 K, with a much reduced PDH structure, the application of a magnetic field reduces the CP while mainly filling the dip. In conventional BCS superconductors, the spectral weight composing the CP will all be transferred from the subgap region [11]. In high-T_c superconductors, however, a large depletion of the spectral weight already exists in the normal state below T^* [1]. Thus, upon onset of superconductivity, the condensation of the quasiparticles in the Fermi surface near the $(\frac{\pi}{2}, \frac{\pi}{2})$ point [19], corresponding to the low-bias subgap region in the tunneling spectra, is a relatively minor contribution to the CP. The spectral weight forming the CP below T_c is mainly contributed from the bias region above the CP position. The feature of the spectral-weight redistribution, along with the suppressed coherence, in high H fields is in clear contrast to previous observations in Bi-2212 [7,20], where no spectral transfer was reported either in the subgap region or in the dip region, despite the significantly suppressed CP caused by H fields.

As Fig. 4(c) shows, above T_c, the CP completely disappears, leaving only the PG feature with a gradual depletion of the DOS. In this case, no change is detected in the spectral weight distribution in fields up to 6 T. This field dependence is in agreement with the previous results from ITS measurements.

III. CONCLUSION

In conclusion, using our ITS, we confirmed the dominant antinodal c-axis tunneling of quasiparticles revealed in the U-shaped spectra. Upon the onset of superconductivity in high-T_c materials, the spectral weight is transferred from the background hump to the CP, generating a dip for a significant transfer at biases slightly above the CP position. In this sense, the dip actively participates in the revelation of the superconductivity. This anomalous transfer from the dip to the CP may originate from the basic mechanism of high-T_c superconductivity.

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