Discriminating the Superconducting Gap from the Pseudogap in $Bi_2Sr_2CaCu_2O_{8+\delta}$ by Interlayer Tunneling Spectroscopy

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Tunneling spectroscopy using a very thin stack of intrinsic Josephson junctions has revealed that the superconducting gap is definitely different from the pseudogap in the Bi₂Sr₂CaCu₂O_{8+ δ} system. In the underdoped region, the conductance peak arising from the superconducting gap is independently observed in the *dI/dV-V* curve and its position is much lower than that of the pseudogap. Near the optimum doping level and in the overdoped region, both peaks are located in close proximity. These findings are in conflict with a previous understanding of the pseudogap.

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It is now widely accepted that the electronic density of states in high- $T_{\rm c}$ superconductors undergoes a decremental change in its spectrum around the Fermi level below a certain temperature T^* , which is much higher than T_c . This change, called the pseudogap (PG) evolution [1], is observed almost commonly in high- T_c superconductors. Since it is believed that PG is closely related to the pairing mechanism of high- T_c superconductivity, the origin of PG and its relation to the superconducting gap (SG) have been attracting wide and continued interest both theoretically and experimentally. Angle resolved photoemission spectroscopy (ARPES) experiments [2] revealed that PG has the same *d*-wave symmetry as SG. Scanning tunneling spectroscopy (STS) experiments [3] suggested that SG smoothly connects with PG at T_c with a sizable magnitude. These results have postulated a picture that the order parameter amplitude persists up to T^* high above T_c , while the macroscopic phase coherence sets in only below T_c [4,5].

While this picture appears persuasive, the *T* dependence of SG [6], penetration depth (λ^{-2}) [7], and the maximum Josephson current [8–10] also seem to indicate the disappearance of the order parameter amplitude at T_c . If SG starts to evolve at T_c , PG must be interpreted differently such as a spin excitation of a certain kind. Thus the elucidation of detailed behavior of SG and PG near T_c is crucially important. In this Letter, we report the results of tunneling spectroscopy (TS) intended to discriminate SG from PG.

In order to obtain a sufficient energy resolution and a clear energy structure in TS measurements, we employed superconductor-insulator-superconductor-type intrinsic Josephson junctions (IJJ) of Bi₂Sr₂CaCu₂O_{8+ δ} (BSCCO) [11]. The most important advantage of the use of IJJ for TS is that we can ascertain the *T* dependence of the *c*-axis resistivity ρ_c of the very portion to be probed. With this means, we can estimate the doping level almost exactly [12]. We have measured more than 20 specimens with different doping levels. From the results, we have deduced the systematic behavior of SG and PG. The major conse-

quence is that SG (order parameter magnitude) disappears at T_c and is definitely distinct from PG.

Specimens used for the interlayer TS are very thin IJJ stacks made of BSCCO crystals with different doping levels. They were fabricated by forming a 10 or 20 μ m square 15 to 20 nm thick mesa (approximately ten junctions connected in a series) on a cleaved surface of a BSCCO crystal grown by the traveling-solvent-floating-zone method [12] or by the self-flux method. Before the photolithograph process, a 25 nm thick Ag thin film and a 50 nm thick Au thin film were evaporated on the cleaved surface and then annealed at 430–450 °C for 1 to 1.5 h in oxygen atmosphere or in vacuum, depending on the carrier doping level. The other fabrication processes were detailed in previous publications [8,13].

Current-voltage (*I-V*) characteristics were measured by the short pulse method [14] with 1 μ s wide current pulses at a duty of 0.05%. The influence of heating due to selfinjection of current is less than 3% on the voltage scale for 20 μ m square overdoped specimens, as detailed elsewhere [14]. Since smaller junction size reduces the heating, the influence of heating is expected to be much less than 3% for 10 μ m square junctions and for underdoped specimens, in particular. In this method, the maximum applied voltage was approximately 1.5 V. When the pulse voltage was higher, the specimens were destroyed by the electric field surrender during measurements. This limited the voltage range to less than approximately 120 mV for a single junction.

Figure 1 shows ρ_c -*T* characteristics for four specimens with different doping levels. It is known that in the BSCCO system there is a clear and nearly unique relationship [12] between the doping level δ and the ratio $r = \rho_c^{\text{max}}/\rho_c^{300 \text{ K}}$, where ρ_c^{max} is the maximum ρ_c just above T_c and $\rho_c^{300 \text{ K}}$ is ρ_c at 300 K. The relationship is expressed as $\delta = 0.174 + 0.321/(r + 1.932)$ with an error of $\Delta \delta = \pm 0.005$. Thus, the doping levels of the specimens in Figs. 1(a)-1(d) were estimated as $\delta = 0.22$ (underdoped), 0.24, 0.25 (near optimum), and 0.28 (overdoped),



FIG. 1. Temperature dependence of the *c*-axis resistivity calculated from the stack resistance R_c for four specimens having different carrier doping levels: (a) $\delta = 0.22$ (underdoped), (b) $\delta = 0.24$, (c) $\delta = 0.25$ (near optimum), (d) $\delta = 0.28$ (overdoped). The dashed line in (c) is the contact resistance inferred from the oscilloscope *I*-V measurements.

respectively, as indicated in Fig. 1 together with a value for $T_{\rm c}$.

The inset of Fig. 2 displays an oscilloscope image of the *I-V* characteristics for the specimen of Fig. 1(c), whose doping level is near the optimum. The characteristics exhibit multiple resistive branches, from which we determined the value of N = 13 for the number of junctions. The maximum Josephson current is rather homogeneous except for one junction which is probably located in contact with the Ag/Au electrode on the top. Since the *I-V* characteristics are measured for all the junctions in series, the influence of the outermost junction is negligible. The



FIG. 2. I-V characteristics at different temperatures for the specimens in Fig. 1(c). The inset displays the oscilloscope I-V characteristics measured at 6 K, in which the lower branch is used for the tunneling spectroscopy.

numbers of junctions determined in this way for the specimens in Figs. 1(a)-1(d) are 12, 7, 13, and 14, respectively. The main panel of Fig. 2 shows the *I*-*V* characteristics measured by the short pulse method for the same specimen. In this *I*-*V* curve, the gap position is not necessarily clear unlike the case of overdoped specimens [6]. Furthermore, the gap structure in the *I*-*V* curve is much less discernible for underdoped specimens.

Figures 3 show four sets of dI/dV-V characteristics obtained numerically from the short pulse *I*-V characteristics for the same specimens shown in Fig. 1 at various temperatures from 10 to 180 K. The values for V are for a single junction. The thick curves indicate the results obtained at a temperature very close to T_c . Each curve is shifted vertically for an appropriate amount for convenience. For the specimen in Fig. 3(a), the excess conductance at V = 0 V is approximately 4.8% of the dI/dV value at 180 K and V = 100 mV. Those for specimens in Figs. 3(b) to 3(d) are 8.4%, 3.4%, and 12%, respectively. Thus, the influence of the excess conductance on the electronic density spectra seen in Fig. 3 is regarded as very small.

When we turn to dI/dV-V curves for $T > T_c$ in Fig. 3, it is clearly seen that they exhibit a significant gap structure even above T_c . This strongly T dependent structure is presumed to be due to a PG observed for the BSCCO system extensively by various methods, particularly by ARPES [2,5] and STS [3,15]. Then, the dI/dV-V curves indicate that PG starts to evolve at a higher than 200 K temperature for specimens in Figs. 3(a)-3(c) (underdoped and optimum), while it starts to evolve at 140 K for the specimen in Fig. 3(d) (overdoped). The PG magnitude is basically reflected by the depth and the width of the dI/dV-V curve, e.g., at $T_{\rm c}$ (thick line). It is evident from Fig. 3 that the PG magnitude systematically decreases as the doping level increases from Figs. 3(a) to 3(d). This tendency is consistent with the previous observations by ARPES and STS. In the present dI/dV-V characteristics of Figs. 3(a) and 3(b), the PG peak is located outside the maximum voltage due to the experimental limitation. Therefore, we approximately estimated the peak position by fitting the BCS model [15] with a finite quasiparticle relaxation time to the dI/dV-Vcurve near $T_{\rm c}$ (thick line). The estimates of the peak position are 150 \pm 20 mV, 130 \pm 20 mV, 80 \pm 10 mV, and 30 ± 5 mV, for Figs. 3(a) to 3(d), respectively. The PG magnitude decreases with the doping level δ but still persists in the overdoped region. This is in accordance with the existence of ρ_c upturn, which is clearly present in ρ_c -T curves in the overdoped region.

In Fig. 3(a), the dI/dV-V curve at 10 K exhibits a small peak at around V = 80 mV, which changes from a small peak to a cusp as T increases and disappears at T_c . This peak is superposed on the shoulder of the PG conductance peak, so that the T dependence of the peak position is not very clear. However, a close inspection reveals that it tends to shift toward lower energies as T increases. We observed a similar peak and behavior for all the specimens



FIG. 3. dI/dV-V curves at various temperatures for the specimens in Fig. 1. Each curve is shifted vertically for an appropriate value. The thick lines indicate the curve very close to T_c . The normal tunneling resistance for the specimen in (d) exhibited a characteristic T dependence very similar to those in Ref. [6].

that were measured with a similar δ value. Based on these observations, we can conclude that the peak near V = 80 mV corresponds to SG. The SG magnitude $2\Delta_{pp}$ defined as half the peak separation at 10 K is 79 meV, which is also reasonably compared with the STS results [3,15,16].

The present result implies that SG disappears at T_c and PG which exists above T_c is distinct from SG. Figures 3(a) and 3(b) also indicate that the PG magnitude is much greater than SG in the underdoped region. The relationship between SG and PG is rather different from the ARPES and STS results in which SG connects smoothly at T_c with PG. It appears that the smooth connection of both gaps reflects the behavior only to be observed near the opti-

mum doping, as seen in Fig. 3(c). The present results also imply that the quasiparticle excitation spectrum has two energy scales. This is also argued in the underdoped YBa₂Cu₃O_{7-x} [17–19] and La_{2-x}Sr_xCuO₄ [20,21] through different experimental probes.

Figures 3(a)–3(d) show that, as the doping level increases, the superconducting peak becomes increasingly pronounced, the height becomes greater, and the peak sharper. The behavior reflects that the superfluid density increases significantly as the doping level increases from the underdoped to overdoped region. With these changes in the peak profile, 2Δ remains almost unchanged from $2\Delta_{pp} = 79$ meV at $\delta = 0.22$ in the underdoped region to



FIG. 4. *T* dependence of $2\Delta_{pp}$ for SG and, partly, for PG for the same specimens in Fig. 3. Open squares and triangles indicate PG values and the other symbols indicate SG peak-to-peak values.

 $2\Delta_{pp} = 77$ meV near the optimum level of $\delta = 0.25$. It then decreases to $2\Delta_{pp} = 45$ meV at $\delta = 0.28$ in the overdoped region. In the overdoped region, the SG peak is much sharper than in the underdoped region and the *T* dependence of $2\Delta_{pp}$ is clearly visible. Figure 4 shows the *T* dependence of $2\Delta_{pp}$ for the four specimens together with values for the PG peaks that are visible. It is noted that the PG values for $\delta = 0.28$ obtained by fitting are much smaller than the $2\Delta_{pp}$ values.

It should be noted that the PG peak position also shifts to lower energies with the increasing doping level at a faster rate than $2\Delta_{pp}$. Near the optimum doping level, SG and PG are located in rather close proximity. In this situation, both peaks are merged to form a single peak as Fig. 3(c) reflects. It can then occur that SG appears to connect smoothly with PG at T_c near the optimum doping level. Indeed, in that doping range, the characteristics are very similar to the STS results [3,15].

In the overdoped region, the PG peak is less pronounced and the SG peak is dominant so that we observe rather conventional behavior for the evolution of SG. The *T* dependence of the peak position is not necessarily the same as, but comparable to the BCS *T* dependence of the order parameter which disappears at T_c .

In spite of many STS measurements of the BSCCO system, reports on the dI/dV-V curve similar to Figs. 3(a) or 3(b) are astonishingly rare. Oda *et al.* [16] observed a similar dI/dV-V curve for an underdoped BSCCO crystal, although they attributed the shoulder of the pseudogap to a large excess conductance which happened to accom-

pany their characteristics. The rareness of such data might be due to an unknown shift of the oxygen doping level towards the optimum level, which might occur in the doping dependence measurements by surface spectroscopy.

Concerning the origin of PG, several models were proposed [4,22,23], among which the fluctuation model might be in conflict with the present result, since SG evolves independently from PG with a different gap energy. This is at variance with the argument that the tunneling probes the single particle excitation and not the phase coherence and that PG is the order parameter amplitude. The present result is also at variance with a model which assumes the establishment of the macroscopic phase coherence at T_c for bosons that are formed below T^* , which is much higher than T_c . It is also in conflict with a model which assumes a smooth connection of SG and PG.

After the submission of this manuscript, we noted a paper by Krasnov *et al.* [24] with the same conclusion.

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