Interplay between Superconducting and Pseudogap States Revealed by Heating-Compensated Interlayer Tunneling Spectroscopy on Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$

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(Dated: February 8, 2020)

The heating-compensated interlayer tunneling spectroscopy is performed on stacks of Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ intrinsic junctions. The high-accuracy spectra without the local heating, for varying temperatures and magnetic fields, reveal that the spectral weight forming the superconducting coherence peak is mainly contributed from the dip position of the hump structure of the pseudogap state. The observed U-shaped subgap structure is consistent with the weighted antinodal tunneling between Cu-O double layers in a junction as suggested in Ref. 17.

PACS numbers: 74.72.Hs, 74.50.+r, 74.25.Fy

Since the discovery of high-$T_c$ superconductivity in cuprates, various anomalous behaviors have been discovered beyond the standard BCS theory. One of them is the pseudogap (PG) observed in the normal state of the materials, which has aroused a great deal of controversy about the relationship between the PG and the superconducting state [1]. The dispute has been focused on two points. One scenario argues that the PG is a precursor to the superconducting state. The other, however, considers it only as a competing or coexisting order with the superconductivity.

Recently, a variety of powerful measurement tools are developed to study the PG state as well as the superconducting state. The surface probes like the angle-resolved photoemission spectroscopy (ARPES) and the scanning tunneling spectroscopy (STS) on Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ (Bi-2212) single crystals have suggested that the PG is the paired superconducting state [2]. More recent STS studies, however, have shown that the PG is a competing or coexisting order with the superconductivity on the same Fermi surface [3]. The surface probes like the bulk tunneling properties of intrinsic junctions [4] imbedded in single crystals, also seems to reveal that the two states are separate in their physical origins, as manifested by the peak-dip-hump (PDH) structure below the superconducting critical temperature $T_c$ [5]. The PDH characteristics in the ITS used to be considered as the superposition of the superconducting coherence peak (CP) and the PG hump structure below $T_c$.

The onset of the superconductivity in cuprates is accompanied by the emergence of a sharp CP that forms the gap edge on the background of pre-depleted quasiparticle DOS near the Fermi surface. Clearer distinction between the CP and the hump reveals in an external magnetic ($H$) field applied along the $c$ axis. An $H$ field, a tuning parameter of the phase coherence, reduces the spectral weight in the CP and redistributes it in a fashion corresponding to the state of lower condensation. As in the BCS case [6], the total spectral weight is expected to be conserved in the process. Existing ITS measurements in Bi-2212 near $T_c$, however, revealed that, as the CP reduces significantly in $H$ fields, the spectral weight in the subgap (the bias region below the CP) remains almost unaltered or even decreases [7, 8]. Since, in these studies, the spectral weight of the hump was not altered in the process, either, the results imply that the total spectral weight in varying fields is not conserved, which is hard to be accepted. The delicate transfer of the relative spectral weight between the CP and the hump, which can be a key to understanding the basic mechanism of the high-$T_c$ superconductivity, is thus yet to be clarified accurately.

In this letter we report the detailed PG behavior both in the normal and the superconducting states and analyze the interplay between the PG and the superconducting states, on the basis of the temperature ($T$), the magnetic field ($H$), and the doping ($p$) dependencies of the spectrum. Recently, it has been found that the ITS is susceptible to the self-heating caused by a finite bias in combination with the poor thermal conductivity of the Bi-2212 [9]. To eliminate the self-heating, we adopted the heating-compensated constant-$T$ ITS on stacks of both overdoped and underdoped Bi-2212 intrinsic junctions [10]. Using the bona-fide electronic spectra from our improved ITS the transfer of the spectral weight was traced with high accuracy in varying $H$ fields, which leads to clarifying the origin of the PDH structure and the relation between the PG and the superconductivity. In addition, the pronounced U-shaped subgap spectra observed in our ITS constitute another distinctive feature from the existing ITS results, providing an insight into the nature of the interlayer tunneling.

The Bi-2212 single crystals were prepared by the solid-state-reaction method (UD75, OD88) and the traveling-solvent-floating-zone method (UD77) [11]. Instead of a usual mesa structure, a sample was prepared into a 3×3 μm$^2$ stack of intrinsic junctions sandwiched between two Au-film electrodes, by employing the double-sided-cleaving of Bi-2212 crystals, photo-lithographic micropatterning, and ion-beam etching [12]. A sample consisted of two stacks [the inset of Fig. 1(a)]; the left one represents the sample stack on which the ITS was taken. The right one, placed a fraction of micrometer apart from the sample stack and consequently in strong inter-stack...
thermal coupling through the common bottom Au electrode, is for the in-situ thermometry of the sample stack. The proportional-integral-derivative control of the constant sample temperature during the ITS in a finite bias is described in detail in Ref. [10]. The temperature during a sweep was kept constant within ~0.1 K for a bias per junction up to ~120 mV (~70 mV, ~40 mV) for UD75 (UD77, OD88). Measurements were made along the highest-bias quasiparticle curve, i.e., the last branch. The $dI/dV$ curves were obtained using the lock-in technique operating at 33.3 Hz.

The c-axis superconducting transition temperatures $T_c$ were 75.2 K (UD75), 77.0 K (UD77), and 88.3 K (OD88). The corresponding doping levels determined by the empirical relation [3], $T_c=95[1-82.6(p-0.16)^2]$, were $p=0.109$, 0.112, and 0.19, respectively. The underdoped(UD75 and UD77) and overdoped (OD88) sample stacks contained $N=15$, 24, and 19 intrinsic junctions, respectively, as determined by the number of quasiparticle branches in the zero-field $I$-$V$ curves at 4.2 K (not shown).

A series of interlayer tunneling spectra $dI/dv(v)$ of OD88 and UD77, for varying $T_c$, are displayed in Fig. 1, where the voltage is normalized by the number of junctions as $v=V/N$. In the normal state of both samples above $T_c$ the low-bias DOS is smoothly depleted, revealing the PG. At a $T_c$ below $T_c$ a sharper peak (the CP) develops inside the PG, constituting the PDH structure. Further lowering $T_c$, the fast sharpening CP with the growing SG size overshadows the spectrum, leaving only the CP apparent. The PG with the hump is more conspicuous in the underdoped UD77. Both samples below $T_c$ show the U-shaped DOS in the subgap region, which is contrasted to the mainly V-shaped DOS observed previously [3]. An interesting observation from all our samples is that all the ITS curves in the PG state above $T_c$ intersect at a single point inside the PG. Similar feature was observed in the ARPES measurements on the antinodal region of the Fermi surface above $T_c$ [12], which may provide an evidence that the interlayer tunneling is dominated by the antinodal hopping. The fluctuating conductance at zero bias sufficiently below $T_c$ in Fig. 1 was caused by the Josephson pair tunneling.

The U-shaped DOS in the subgap region poses an important implication to the interlayer tunneling in high-$T_c$ superconductors. The tunneling quasiparticle current in a Josephson junction is written as $I(V) = 4\pi e \int_{\xi_{\phi}}^{\xi_{\phi}+\Delta_{c}} |T_{k,p}|^2 |f(\xi_{k}) - f(\xi_{p})| d\xi_{k}$, where $T_{k,p}$ is the tunneling matrix element, $\xi_{k} \equiv \sqrt{\Delta_{c}^2 - \Delta_{0}^2}$, and $f(\xi_{k})$ is the Fermi function [14]. Assumed coherent elastic tunneling $|T_{k,p}|^2 = |T_{k,p}^0|^2 \delta_{k,p}$ [15], along with the $d_{x^2-y^2}$-wave gap symmetry and the relation $\sum_{k} = 4(2\pi)^2 \int d\xi_{k}$, leads to

$$I(V) = \frac{1}{2\pi e R} \int_{0}^{2\pi} d\phi |T_{\phi}|^2 \int dEN(\phi, E)N(\phi, E + eV) \times \{f(E) - f(E + eV)\}, \quad (1)$$

where $A$ is the junction area, $\phi \equiv \tan^{-1}(k_y/k_x)$, and $N(\phi, E) = \text{Re} \{(E - i\Gamma)/(E - i\Gamma)^2 - \Delta_{0}^2 \cos^2 \phi)^{1/2}\}$ with the quasiparticle scattering rate, $\Gamma$. The dotted curves in Fig. 2(a) are the numerical result for a $k$-independent isotropic tunneling matrix element $T_{\phi} = 1$ [16] with $\Delta_{0} = 20$ meV and $\Gamma = 0.05$ meV at $T_c = 4.2$ K. On the other hand, the band calculation for a crystal with the tetragonal symmetry predicts that the $c$-axis quasiparticle tunneling dominates near the antinodal points in the first Brillouin zone, satisfying $T_{\phi} = t_{c} \cos^2 2\phi$ [17]. The solid curves in Fig. 2(a) correspond to this

![FIG. 1: (color online) The interlayer tunneling spectra $dI/dv$ for (a) the overdoped OD88 and (b) the underdoped UD77 as a function of bias voltage per junction at various $T_c$. Inset of (a): the measurement configuration.](image)

![FIG. 2: (color online) (a) Calculated $I$-$V$ and $dI/dv$($v$) curves with Eq. (1) for $T_{\phi} = 1$ (dotted curves) and $T_{\phi} = t_{c} \cos^2 2\phi$ (solid curves) at $T_c = 4.2$ K. (b) The $T_c$ dependencies of the SG ($\Delta_{sg}$, open symbols) and the PG ($\Delta_{pg}$, filled symbols).](image)
anisotropic $T_\phi$, which reduces the low-energy quasiparticle tunneling near the nodal points. It leads to the U-shaped tunneling conductance, while sharpening the CP. Thus, the pronounced U-shaped spectra in our ITS (Fig. 1) strongly suggest again that the antinodal hopping is highly weighted in the interlayer tunneling.

The $T$ dependence of the SG energy $\Delta_{sg}$ (open symbols) and the PG energy $\Delta_{pg}$ (filled symbols) is shown in Fig. 2(b), which clearly reveals the coexistence of the CP and the hump. For each doping level $\Delta_{sg}$ reduces as $T$ approaches $T_c$. By contrast, the $\Delta_{pg}$ exhibits a more complicated $T$ dependence. For UD77 and UD75 in the normal state above $T_c$, $\Delta_{pg}$ shrinks with decreasing $T$. But overdoped OD88 shows a local maximum of $\Delta_{pg}$ above $T_c$. The $\Delta_{pg}$ in each doping level reaches a local minimum at a $T$ slightly below the respective $T_c$ and rises with further lowering $T$. In OD88, $\Delta_{pg}$ was observed to grow from zero with lowering $T$ below $T_c$. UD77 and UD75 are expected to exhibit similar behavior, except for more extended temperature scales.

We now focus on whether the dip is caused by the simple superposition of the CP and the PG hump or by the transfer of the spectral weight. As in OD88 and UD77, the dip in the superconducting state in our ITS disappears when $\Delta_{sg}$ becomes comparable to $\Delta_{pg}$ with lowering $T$ below $T_c$. For UD75, however, the PDH structure exists even down to $\sim$20 K because $\Delta_{pg}$ is sufficiently larger than $\Delta_{sg}$ at the temperature. Fig. 3 illustrates the progressive evolution of the simple hump structure into the PDH one in UD75 with decreasing $T$. The CP grows sharply with decreasing $T$ below $T_c$. Along with it, the position of the dip shifts to a higher voltage with deepening its depth. 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 $T$.

The redistribution of the spectral weight is more clearly visible in a high $H$ field, applied in this study up to 6 T along the $c$ axis. As in the case of varying $T$, for a varying phase coherence in an $H$ field, the relative weight of the DOS redistributes between the CP and the hump. Figs. 4(c) and (d) [Figs. 4(g) and (h)] display the $H$-field dependence of the spectra for UD75 [OD88] at 65 and 55 K [82.6 and 80 K], respectively. The insets of Figs. 4(d) and (g) [Fig. 4(e)] show the total spectral weight, obtained by integrating the respective curve over the corresponding zero-field [79 K] value. The cutoff voltage $v_c$ of integration was selected well above the merging position of the different-$H$ and different-$T$ spectra around the hump. The resulting virtually $H$- and $T$-independent total spectral weight is consistent with the sum rule and, thus, confirms the reliability of our description for the transfer of the spectral weight based on our ITS.

The heat-compensated constant-$T$ ITS enabled the accurate tracing of the spectral weight transfer by any external parameters changing the coherence, such as $T$ and $H$. The spectra of UD75 in Fig. 4(d), at a fixed $T$ of 55 K, reveal that the CP is suppressed rapidly while the dip fills up with increasing $H$ field. Similar trend holds at 65 K [Fig. 4(c)], although the CP and the corresponding spectral change are much less pronounced. This trend also holds for the overdoped OD88 with a smeared PDH structure as in Figs. 4(b) and (g). In comparison, in the process, only a small portion of the CP spectral weight is transferred to the subgap region, especially for UD75. In conventional BCS superconductors the spectral weight composing the CP would be all

![FIG. 3: (color) Color-coded plot of the interlayer tunneling spectra per junction for UD75 for varying $T$. The solid curves are the measured spectra, with the edge of $\Delta_{pg}$ denoted by * symbols.](image)

![FIG. 4: (color) (a) and (e): zero-field spectrum change for sets of temperatures for UD75 and OD88. (b) and (f): the corresponding curves in $H=6$ T, almost representing the PG structure at given $T$ for the two samples. (c), (d), (g), and (h): $H$-field dependence of the peak-dip-hump structure for the corresponding $T$. Insets: the total spectral weight vs $T$ [(e)] and $H$ field [(d) and (g)].](image)
transferred from the subgap region. In high-$T_c$ superconductors, however, large depletion of the spectral weight in the Fermi surface already exists in the normal state below $T^*$ [1]. Thus, upon onset of the superconductivity the condensation of the quasiparticles is limited to the Fermi arc in the vicinity of $(\pi, 0)$ point [12], corresponding to the low-bias subgap region in the tunneling spectra, which makes a relatively minor contribution to the CP. Recent ARPES measurements indicate that the Fermi arc extends for higher doping [22], allowing more quasiparticles to condense to form the CP along with.

That may explain why in Fig. 4, as the superconducting coherence is enhanced, a higher portion of quasiparticle spectral weight below the crossing points (denoted by the arrows) is transferred to the CP in OD88 than in UD75.

The $T$ dependence of the spectral shape in Figs. 1(a) and (b) as well as Fig. 3 appears to suggest that rapid growing of the CP with decreasing $T$ below $T_c$ is caused by the fast depletion of the spectral weight below the gap as the superconductivity sets in. The trend is more clearly seen in the comparative illustration of the zero-field DOS for a couple of temperatures below $T_c$ for UD75 [Fig. 4(a)] and OD88 [Fig. 4(e)]. For UD75, as $T$ decreases from 65 K to 55 K, the spectral weight transferred below the gap [Region (1)] and in the dip [Region (3)] constitutes the CP [Region (2)] and the hump [Region (4)]. For OD88, similar transfer of spectral weight takes place from Regions (1) to (2) as $T$ lowers from 82.6 K to 80 K. Although the superconductivity is not completely suppressed in 6 T [as evidenced by the presence of the shoulder near 40 mV of Fig. 4(b)] the spectral curves at different $T$ in Fig. 4(b) and (f) almost represent the PG structure at the respective temperatures [23]. This indicates that the apparent further depletion of the low-energy spectral weight with lowering $T$ [Region (1) of Figs. 4(a) and (e)] effects the transformation of the PG structure itself, accumulating more weight in the Region (2) of Fig. 4(b) (the shoulder region would disappear for higher fields) and Fig. 4(f). Additional spectral-weight change, upon onset of superconductivity, on the background of this PG is then clearly seen in Figs. 4(c) and (d) for UD75, and Figs. 4(g) and (h) for OD88, with the growing CP with lowering $H$ for the temperatures explored. These figures clearly indicate that the spectral weight constituting the CP is mainly contributed by the transfer from the bias region above the CP position rather than from the subgap region, which contradicts to the general perception including the BCS behavior. This is the main finding of this work. 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 [24], where no spectral transfer was reported either in the subgap region [1] or in the dip region [8], despite the significantly suppressed CP by $H$ fields.

In conclusion, using our ITS, we confirmed the dominant antinodal $c$-axis tunneling of quasiparticles as revealed in the U-shaped spectra. Upon onset of the superconductivity in Bi-2212, the spectral weight is transferred from the background PG hump to the CP, generating a dip for a significant transfer at the bias 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.

This work was supported by KOSEF through the National Research Laboratory program. We are grateful to N. Momono, M. Oda, and M. Ido in Hokkaido University, Japan, for providing the underdoped single crystals for UD77 and valuable communications.

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