

Interlayer Tunneling Spectroscopy for Slightly Overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$

Minoru Suzuki,^{1,*} Takao Watanabe,² and Azusa Matsuda²

¹*NTT Basic Research Laboratories, Nippon Telegraph and Telephone Corporation,
162 Tokai, Ibaraki 319-1193, Japan*

²*NTT Basic Research Laboratories, Nippon Telegraph and Telephone Corporation, 31 Morinosato,
Wakamiya, Atsugi, Kanagawa 243-0198, Japan*

(Received 4 February 1999)

We have measured the interlayer tunneling characteristics of slightly overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ in the c -axis direction from 10 to 220 K by using very thin mesas containing approximately ten CuO_2 double layers. The superconducting gap 2Δ is 50 meV at 10 K and shows a temperature (T) dependence similar to that of the BCS theory. A pseudogap evolves below 150 K, where the c -axis resistivity ρ_c is semiconducting. The normal-state tunneling resistance R_N shows a linear T dependence down to T_c , exhibits an abrupt decrease at T_c , and saturates at low T . This reflects scattering arising from an electronic interaction that is relevant to the high- T_c superconductivity. [S0031-9007(99)09456-9]

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

It is probably an essential feature of high- T_c superconductors that carriers are mostly confined within quasi-two-dimensional (quasi-2D) metallic sheets (CuO_2 double layers), while the sheets are weakly coupled by the Josephson effect. Within these quasi-2D CuO_2 layers, carriers are believed to behave as different entities, such as holons and spinons [1]. In such a case, the carrier transport mechanism parallel and perpendicular to the CuO_2 layers is expected to be different. Since this picture is closely related to the occurrence of high- T_c superconductivity in some models [2], it is particularly important to gain an understanding of the interlayer transport and relevant electronic states. However, no general consensus has yet been reached even as to the semiconductive temperature (T) dependence of the c -axis resistivity ρ_c [3].

In some highly anisotropic high- T_c superconductors, the layered crystal structure forms a stack of Josephson tunnel junctions [4,5]. If such tunnel junctions are properly handled, they provide a unique means for interlayer tunneling spectroscopy, which directly probes the c -axis transport and simultaneously the relevant electronic states. This technique is different from photoemission spectroscopy or scanning tunneling spectroscopy (STS) in that the former probes the energy of carriers traversing the layers, while the latter deals only with the electrons extracted into a vacuum from the outermost CuO_2 layer. Furthermore, interlayer tunneling spectroscopy employs superconductor-insulator-superconductor (SIS) junctions, which offer better energy resolution than superconductor-insulator-normal-metal (SIN) junctions.

For the interlayer tunneling spectroscopy experiments, we fabricated very thin small mesas containing approximately ten junctions on the surface of a $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ crystal by using fine processing technology. We employed these mesas to measure the tunneling characteristics of slightly overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ crystals by the short pulse method. The influence of heating caused

by quasiparticle current injection was negligible in the observed current-voltage (I - V) characteristics. From these characteristics, we found that the magnitude of the superconducting gap 2Δ is approximately 50 meV for these crystals. We also observed the evolution of a pseudogap below 150 K with a magnitude ranging from 37 to 62 meV. The most notable observation in this study is the characteristic T dependence of the normal state tunneling resistance R_N , which behaves differently below T_c . This not only suggests that an electron-electron interaction is relevant to high- T_c superconductivity but also provides the temperature (T) dependence of the phase relaxation time τ of the scattering related to the electronic interaction.

We fabricated mesas about 15 nm thick using conventional photolithography and the Ar ion milling technique. $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ crystals were grown by the traveling-solvent-floating-zone method [6] and annealed at 600 °C for 100 h. Prior to the photolithography, a 75 nm thick Ag/Au bilayer was evaporated on the cleaved surface of the crystals. Then the crystals were annealed at 430 °C in an oxygen atmosphere to reduce the contact resistance. The mesa thickness was controlled by the ion-milling time so that each mesa contained ten Josephson junctions. Eventually, the mesas contained nine to eleven junctions, which we confirmed from the number of resistive branches in the I - V curve. The mesa size was typically $20\ \mu\text{m} \times 20\ \mu\text{m}$. A 250 nm thick SiO film was evaporated in a self-aligned manner to provide insulation. A 350 nm thick Au layer was evaporated onto the mesa surface for the upper electrode wiring. The samples had a three-terminal configuration. The contact resistance of the mesas was typically 0.2–0.3 Ω and negligible in the present tunneling measurements. The other fabrication details have been described elsewhere [7].

The mesa resistance R_c showed a metallic T dependence down to 140 K and semiconductive behavior from 140 K to $T_c = 87.1$ K, where R_c showed a sharp resistive

transition. The value for ρ_c at 300 K was estimated from R_c to be $32.5 \Omega \text{ cm}$. As regards the oxygen content δ , we obtained an estimate of $\delta = 0.28$ by comparing the R_c - T curve with the ρ_c - T curves for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystals with a known oxygen content [6]. The mesas were, therefore, slightly overdoped.

Previously used $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ mesas have usually been thick and have contained a large number of Josephson junctions [5]. The tunneling measurements made on them were, to a greater or lesser degree, accompanied by heating due to current self-injection. In an extreme case, a large current injection drove the specimen into the normal state [5]. Although the heating was greatly reduced by reducing the mesa thickness, the influence of heating was still noticeable even in such very thin mesas and caused a reduction in the superconducting gap voltage [7,8]. It also caused ambiguities in the local temperature of the sample and the characteristic energy value. Thus we needed further reduction in heating. Therefore, we adopted the short pulse method [9], in addition to using very thin mesas. In this method, current pulses $1 \mu\text{s}$ wide and with a duty of 0.02% were supplied from an arbitrary waveform generator. The output voltage was measured with a four-channel digital oscilloscope at 500 ns from the rise of a pulse. The voltage was determined by averaging 50 accumulated data values. We also estimated the influence of heating induced by the current injection by measuring the voltage response shape. The voltage response had a peak and a subsequent decay within 500 ns of the pulse rising. The decay corresponds to the influence of the heating. We found that the magnitude of the voltage decay reached a maximum of 2.6% of the total height when the current pulse height I_p was 80 mA. When $I_p > 80$ mA, the voltage decay became less significant. Thus the influence of heating on the gap parameter is less than 3% in the present tunneling measurements.

The inset in Fig. 1 shows an I - V curve at 10 K for a mesa containing eleven junctions. The curve exhibits resistive branches with a spacing of approximately 25 mV, implying that the mesa contains eleven junctions in series. Figure 1 shows a set of I - V curves for this mesa measured by the short pulse method at various temperatures from 10 to 220 K. In the measurements, we applied a magnetic field of 1 T parallel to the c axis to suppress the Josephson current [10]. For clarity hereafter, the voltages are divided by the number of junctions ($N = 11$). Thus the voltages in the figures indicate the single junction value. In Fig. 1, the superconducting gap structure is clearly seen in each curve below T_c . No negative resistance can be observed in these curves, indicating that the heating caused by the current injection was greatly reduced as expected. The T dependence of the I - V curves reflects a systematic decrease in 2Δ with decreasing T . Another feature of these I - V curves is a characteristic T dependence of R_N , which is described in greater detail below.

Figure 2(a) shows a set of dI/dV - V curves below T_c which we obtained numerically from the data in Fig. 1.

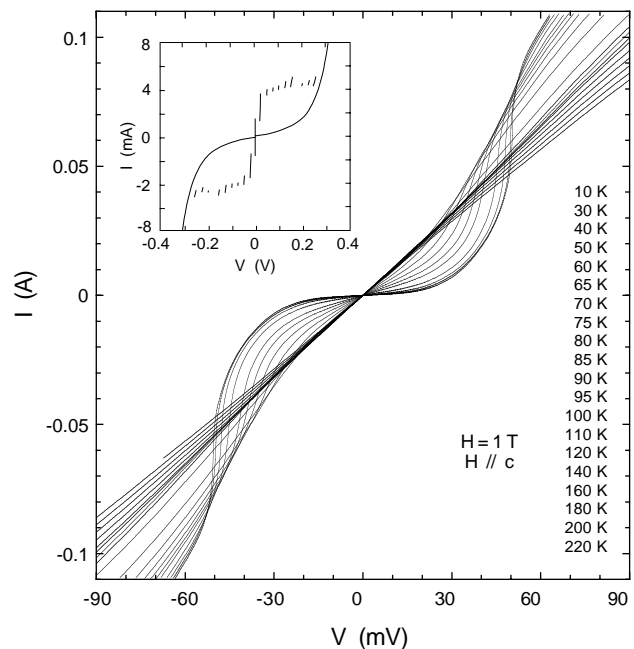


FIG. 1. The I - V characteristics of a $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ mesa containing 11 Josephson junctions measured with $1 \mu\text{s}$ current pulses at temperatures from 10 to 220 K. A magnetic field parallel to the c axis was applied to suppress the Josephson current. The inset is an oscilloscope trace of the I - V curve in the absence of a field, showing eleven resistive branches separated by a space of 25 mV.

In the calculation, we used least-square smoothing. The small dI/dV values at $V = 0$ indicate slight leakage conductance for the junctions in the mesa. The systematic change in the dI/dV curves clearly indicates that 2Δ decreases with increasing T . As an approximation to 2Δ , we use $2\Delta_{pp}$ defined as half the separation of the peaks in the dI/dV - V curves. Thus we obtain values of $2\Delta_{pp} = 50$ meV at 10 K and $2\Delta_{pp} = 30$ meV at 80 K, and $2\Delta_{pp}$ appears to vanish when T approaches T_c . In the present study, we obtained values of $2\Delta_{pp} = 48$ to 53 meV at 10 K for different samples. These values are smaller than the STS result reported by Renner *et al.* [11] by a factor of 0.8–0.9. The dI/dV values are nearly proportional to V^2 for $V < 40$ mV at low temperatures, which is consistent with the d -wave symmetry of the order parameter.

Figure 2(b) shows a set of dI/dV - V curves at various temperatures from 85 to 220 K. The curves at 90 K and above form a broad peak structure, providing evidence for the evolution of a pseudogap in slightly overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$. This structure continues to exist up to 150 K, indicating that the pseudogap evolves below approximately 150 K. This pseudogap structure is similar to that previously observed for a $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ mesa [8] except that it has a much greater energy value, which resulted from the significant reduction in heating in the present study. Figure 2(b) reflects only an obscure relationship between the superconducting gap and the pseudogap. It is not

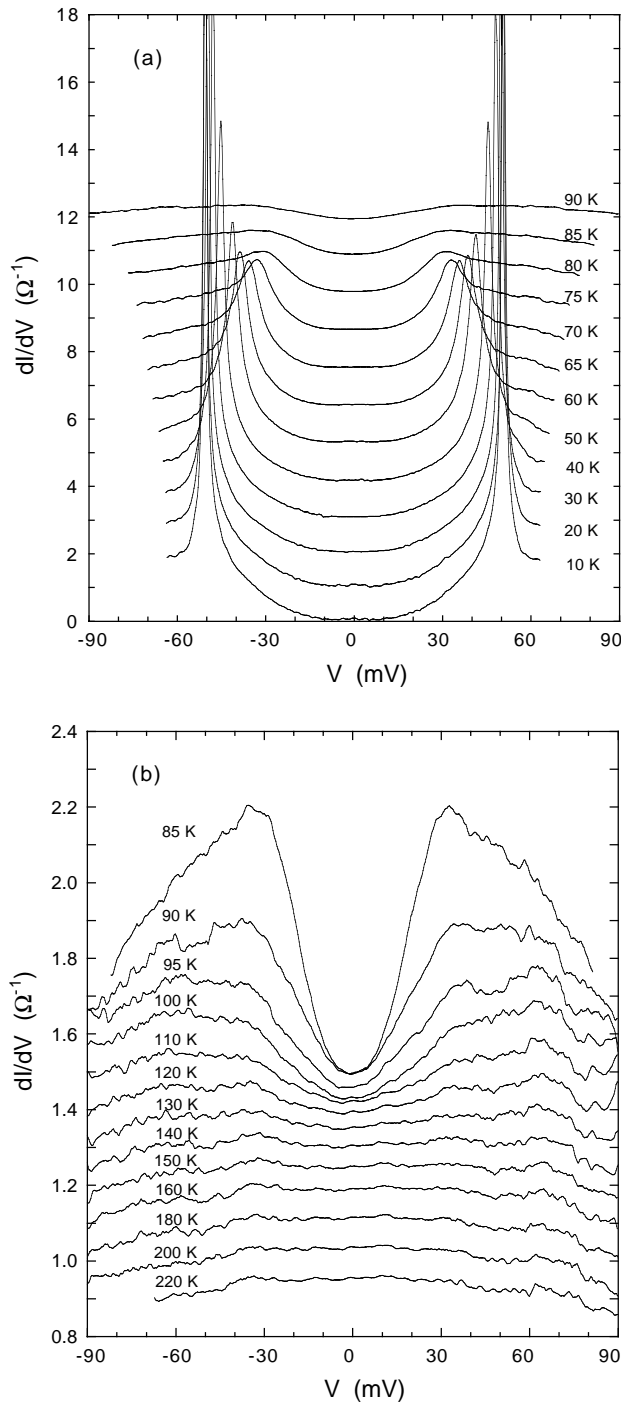


FIG. 2. (a) dI/dV - V curves below T_c obtained numerically from the data in Fig. 1. Each curve is shifted by $1 \Omega^{-1}$ to make it easier to see. (b) dI/dV - V curves above T_c obtained numerically from the data in Fig. 1. Each curve is shifted by $0.05 \Omega^{-1}$.

necessarily clear from this figure that the superconducting gap evolves within the pseudogap.

While the view of a single broad peak appears valid for the dI/dV spectrum, a closer inspection of the peak structure reveals two broad peaks centered at 37 and 62 mV. This structure can be observed in different samples and is probably one of the intrinsic properties of

this system. An interpretation is that the peak at 37 mV corresponds to the pseudogap while the peak at 62 mV arises from a band-structure effect such as a van Hove singularity. This peak at 62 mV appears to exist below T_c . In the dI/dV - V curves from 40 to 80 K in Fig. 2(a), we find similar traces near 60 mV. This is reasonable if the peak at 62 mV is of band-structure origin.

Figure 3 shows the T dependence of $2\Delta_{pp}$ together with the two peak positions. To contrast with the conventional behavior, we depict the BCS 2Δ curve (solid line) in the weak-coupling limit. There is a definite difference in the T -dependent behavior in a higher than 30 K range. Since $2\Delta_{pp}$ becomes larger than real 2Δ with increasing T [12], the difference is actually much greater than that shown in Fig. 3. This implies that 2Δ is much more T dependent in a lower T range than the BCS 2Δ behavior. The significant upward deviation near T_c probably indicates that the superconducting peak is hybridized with the pseudogap peak. We observed a similar T dependence of $2\Delta_{pp}$ for different samples. Some samples differed in the degree of upward deviation near T_c . The broad peaks in the dI/dV - V curves above T_c showed little shift with increasing T , which is consistent with the observation by Renner *et al.* [13].

Figure 4 shows the T dependence of R_N and its large change at T_c . It is known that the thermal conductivity in the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ system is enhanced below T_c [14]. However, its degree of approximately 20% brings about very little change if any in the tunneling result via heating. It is therefore difficult to explain this change in terms of the thermal conductivity anomaly.

The T dependence of R_N , shown in Fig. 4, is the most important result in the present study. This can be obtained only by interlayer tunneling spectroscopy. The dashed line in the figure indicates the T dependence of R_c/N for comparison. The T dependence of R_N is characterized by the following three points. First, R_N shows a linear T

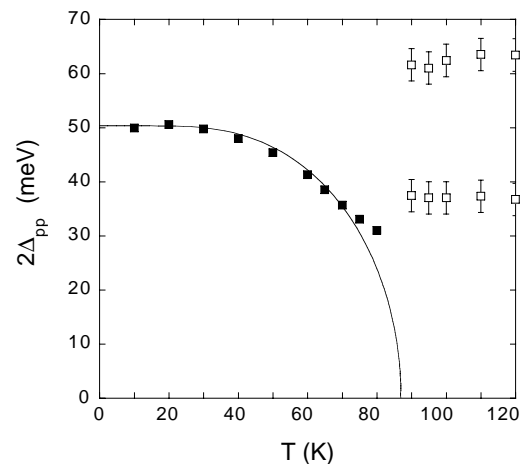


FIG. 3. The T dependence of $2\Delta_{pp}$ defined as half the separation of the dI/dV peaks. The open squares indicate the pseudogap peaks as described in the text.

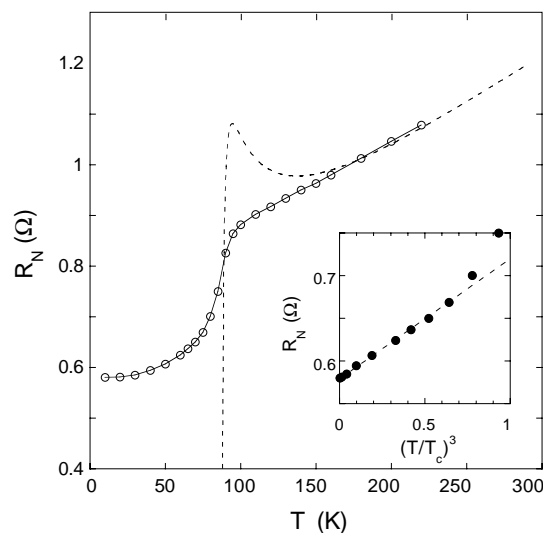


FIG. 4. The T dependence of R_N . The dashed line shows the T dependence of R_c/N . The inset shows the relationship R_N vs T/T_c . The solid line in the main panel and the dashed line in the inset are guides to the eyes.

dependence above T_c . Second, R_N coincides with R_c/N for $T > 160$ K, but deviates from it for $T < 160$ K, where ρ_c is semiconductive. Third, R_N shows a sharp drop at T_c , and saturates at low temperatures. The first point clearly indicates that the tunneling resistance is not reflected by ρ_c . The deviation of ρ_c from R_N at lower temperatures arises from the nonlinearity of the I - V curve due to the evolution of the pseudogap.

In a simple square barrier model, the tunneling probability scarcely depends on temperature. Indeed, R_N is nearly T independent for tunnel junctions made of conventional superconductors [15]. In this sense, the present result for R_N - T presents a sharp contrast with the conventional behavior. In a quasi-2D system, as in the present case, the electron transfer along the c axis occurs via hopping between the layers. In this case, the hopping is reflected by the scattering in the metallic ab planes [16]. Therefore, R_N is proportional to τ^{-1} , where τ is the phase breaking time in the ab plane. Since τ is known as $\hbar/\tau = \alpha kT$ in the higher than T_c temperature range [17], the linear T dependence of R_N reflects the scattering of carriers in the ab planes. This implies coherent (elastic) interplanar tunneling between adjacent layers [18].

In Josephson tunnel junctions made of conventional superconductors, little change is observed in R_N at T_c . When pair formation involves phonons, electron condensation brings about very little change in the phonon spectrum and therefore little change in the scattering time at T_c . In contrast, if the pairing involves bosons arising from an electronic interaction, the boson spectrum changes greatly at T_c because of the condensation of electrons below T_c . In this case, a significant change is expected in the scattering time at T_c . Therefore, the ob-

served large change in R_N at T_c in the present study strongly implies that the pairing is based on an electronic interaction. A number of experiments have already suggested that the pairing interaction is electronic in origin [19,20]. The present result provides another piece of evidence for the electronic origin of high- T_c superconductivity directly from transport measurements.

It is interesting to know the functional form of the T dependence of R_N . As shown in the inset in Fig. 4, we find that the data is best fitted by the form $A + BT^3$ which suggests the relationship $\tau^{-1} = \tau_0^{-1} + T^3$. It should be noted that this T dependence is assumed for the pair breaking time due to spin fluctuation to explain the NMR spin-lattice relaxation experiments [21].

In conclusion, we have measured the tunneling characteristics of a very thin mesa containing eleven Josephson junctions in the c axis direction for a slightly overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ crystal. The observed I - V characteristics show little indication of the effect of heating. We obtained a value of $2\Delta_{\text{pp}} = 50$ meV for the superconducting gap and values of 37 to 62 meV for the pseudogap, which evolves below 150 K. We also found that R_N shows a characteristic T dependence which suggests scattering caused by electronic interaction relevant to the pairing.

*Present address: Department of Electronic Science and Engineering, Kyoto University, Kyoto 606-8501, Japan.

- [1] P. W. Anderson *et al.*, Phys. Rev. Lett. **60**, 132 (1988).
- [2] S. Chakravarty *et al.*, Science **261**, 337 (1993).
- [3] S.L. Cooper and K.E. Gray, in *Physical Properties of High Temperature Superconductors IV*, edited by D.M. Ginsberg (World Scientific, Singapore, 1994), p. 61.
- [4] R. Kleiner *et al.*, Phys. Rev. Lett. **68**, 2394 (1992).
- [5] R. Kleiner and P. Müller, Phys. Rev. B **49**, 1327 (1994).
- [6] T. Watanabe *et al.*, Phys. Rev. Lett. **79**, 2113 (1997).
- [7] K. Tanabe *et al.*, Phys. Rev. B **53**, 9348 (1996).
- [8] M. Suzuki *et al.*, J. Phys. Soc. Jpn. **67**, 732 (1998).
- [9] M. Suzuki, T. Watanabe, and A. Matsuda (to be published).
- [10] M. Suzuki *et al.*, Phys. Rev. Lett. **81**, 4248 (1998).
- [11] Ch. Renner *et al.*, Phys. Rev. B **51**, 9208 (1995).
- [12] R. Meservey and B.B. Schwartz, in *Superconductivity*, edited by R.D. Parks (Marcel Dekker, New York, 1969), Pt. 1, pp. 117–191.
- [13] Ch. Renner *et al.*, Phys. Rev. Lett. **80**, 149 (1998).
- [14] M.F. Crommie and A. Zettl, Phys. Rev. B **41**, 10978 (1990); **43**, 408 (1991).
- [15] B.N. Taylor, J. Appl. Phys. **39**, 2490 (1968).
- [16] N. Nagaosa, Phys. Rev. B **52**, 10561 (1995).
- [17] A.G. Aronov *et al.*, Phys. Rev. Lett. **62**, 965 (1989).
- [18] N. Kumar *et al.*, Phys. Rev. B **45**, 5001 (1992).
- [19] M. Gurvitch *et al.*, Phys. Rev. Lett. **59**, 1337 (1987).
- [20] B. Batlogg *et al.*, Phys. Rev. Lett. **58**, 2333 (1987).
- [21] L. Coffey, Phys. Rev. Lett. **64**, 1071 (1990).