層間トンネル分光による高温超伝導物性の研究

（課題番号　１２６４０３４４）

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はしがき

本報告書は平成12年度および13年度文部科学省科学研究費補助金基盤研究(C)(2)「層間トンネル分光による高温超伝導物性の研究」の研究成果報告書である。研究代表者は平成11年4月に京都大学に赴任し研究活動を開始した。本研究の骨子となる試料は先端エレクトロニクス技術を用いた微細加工技術により作製されるが、着任当初の研究環境は塵埃のおびただしい古い実験室で微細パタン形成は全く困難であったが、学生とともに環境のクリーン化につとめ、平成13年度夏以降、ようやく10ミクロン角のパターン形成が可能となった。これにより本格的な試料作製が可能となり、以後有意義な実験データが得られるようになった。

本研究では、研究代表者が開発した、短パルス層間トンネル分光法を高温超伝導体のエネルギーギャップ構造の観察・解析に応用するとともに、複雑な高温超伝導体のエネルギー構造を一層広い領域で測定できるように層間分光法の改良を進め、高温超伝導体の新しい物性の理解に資する実験結果を得ることを目的とした。その結果、ビスマス系高温超伝導体 $Bi_2Sr_2CaCu_2O_{8+\delta}$ では超伝導ギャップと擬ギャップを明瞭に観察することが可能となり、擬ギャップが超伝導ギャップの延長ではなく二つのギャップは異なるものである結果を得た。これは、擬ギャップの成因および高温超伝導の機構に関する重要な知見である。

また、$La_{1.9}Sr_{0.1}CuO_4$ 超伝導体でも本方法を適用し素子形成の段階まで研究が進展した。さらに、短パルス層間トンネル分光技術においても、パルス波形の改良により分光エネルギー範囲が大幅に改善され、ビスマス系高温超伝導体において、超伝導エネルギーギャップよりも高いエネルギー位置に新しい構造を発見するに至った。以上の成果は、それ自身重要な意義を有するのみならず、今後も、高温超伝導の発現機構の解明に極めて重要な物性を明らかにするものと予想される。

なお、本研究で微細加工に用いた高温超伝導体単結晶はＮＴＴ物性科学基礎研究所で成長されたものである。本研究の重要な部分は単結晶を育成・提供いただいた渡辺孝夫博士および松田梓博士の好意に負っている。

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研究成果

銅酸化物における高温超伝導の発現機構はまだ明らかにはなっていない。モット絶縁体に近接した強い電子相関を示す電子系のスピンと電荷が相互作用して複雑な様相を呈しており、その本質を実験で抽出することは容易ではない。電子系が対を形成して超伝導が発現されることは良いとして、その対形成相互作用が何に因るのか未だに明らかでない。本来、こうした対形成相互作用を明らかにする実験的手法はトンネル分光であるが、高温超伝導体は化学的不安定性を持ち合わせていること、さらに酸化物であるためにその物質の表面被覆酸化物が利用できないことなどのために、トンネル接合形成が極めて困難で、トンネル分光用の接合が得られない。走査型トンネル顕微鏡を用いたトンネル分光の実験も積極的に押し進められているが、表面を探査することのために物質の内部の相互作用を真に抽出可能か厳密には不明である。むしろ、表面分子構造と探針先端原子の間の波動関数空間分布の影響が強い場合さえ考えられる。こうした中で、層状構造の結晶構造を有する高温超伝導体に内在する固有ジョセフソン接合を利用した層間トンネル分光はこうした表面にかかわる一切の影響がない手法として研究代表者らにより提案された。本研究補助金申請の段階において、この方法にはまだ技術的な問題が見られたが、本研究課題の遂行過程においてその問題が一部解決もしくは軽減され、その結果、高温超伝導の理解において重要な知見を得るに至った。特に、超伝導ギャップと擬ギャップが本来異なる物であることをトンネル分光により明確にしたことは本研究の大きな成果のひとつである。本項においてはそうした成果を含め、得られた研究成果を科学研究費補助金申請書の計画に沿って述べる。研究内容の詳細については末尾に関連論文を付してその説明とする。

1．高温超伝導体 $La_{1.9}Sr_{0.1}CuO_4$ エピタキシャル薄膜の成長

高温超伝導体のイントリンシックジョセフソン接合を利用する層間トンネル分光を応用する物質として $La_{1.9}Sr_{0.1}CuO_4$ 系(LSCO)を考え、よりトンネル接合に近い特性を示すことが予測される、異方性の大きい x=0.1 の組成を選び、そのエピタキシャル薄膜成長実験を進めた。この組成は相図の不足ドープ領域に入り、これまでエピタキシャル薄膜成長の検討は十分なされていない。検討の結果、$SrTiO_3$ の(100)面上に基板温度 730-770℃において(001)面 LSCO 薄膜のエピ

タキシャル成長を達成した。薄膜の転移温度 T_c は 29K で抵抗率の温度依存性は金属的な振る舞いを示し、(001)面成長を裏付けている。なお、as-grown では T_c が 12K であるが、成長直後に成長室に酸素を導入することにより Tc は 16K まで上昇し、酸素中 800℃で熱処理することにより T_c は 29K まで上昇することがわかった。

２．高温超伝導体薄膜への Au 電極接触抵抗低減

高温超伝導体 $La_{1.9}Sr_{0.1}CuO_4$ エピタキシャル薄膜を用いて層間トンネル分光実験を行うためにはエピタキシャル薄膜表面上に微小メサを形成する必要がある。トンネル特性の測定には、メサ上部に配置された Au 電極を用いるが、$La_{1.9}Sr_{0.1}CuO_4$ エピタキシャル薄膜との接触抵抗が十分小さいことが必要である。これまで Au 電極を真空蒸着で形成後 600℃で熱処理する方法を用いたが、それではたかだか $10^{-3}\Omega cm^2$ でしかなかった。十分な精度を有するトンネル分光結果を得るためには。$10^{-5}\Omega cm^2$ 以下の低い接触抵抗が必要である。そのため、Au 電極を真空蒸着する直前に $La_{1.9}Sr_{0.1}CuO_4$ エピタキシャル薄膜の電極形成表面に Ar イオンスパッタクリーニング処理を施し、そのまま真空を破らずに Au を蒸着する方法を用い、さらに高速熱処理を施すことより接触抵抗が 1 桁以上低減され、面抵抗率で $10^{-5}\Omega cm^2$ 程度が達成され、微小メサのトンネル特性測定が可能となった。

３．高温超伝導体 $La_{1.9}Sr_{0.1}CuO_4$ エピタキシャル薄膜上への微小メサ形成とトンネル特性

上記高温超伝導体 $La_{1.9}Sr_{0.1}CuO_4$ エピタキシャル薄膜上にスパッタクリーニングおよび高速熱処理法を用いて接触抵抗を低減した Au 電極を形成した後、フォトリソグラフと Ar イオンミリングにより平面寸法 $10\mu m\times10\mu m$ 厚さ 13～14nm の微小メサ構造を作製した。このメサは格子定数で 10 単位分、CuO_2 層で 20 層に該当する。接触抵抗は 0.5～1Ωであり、メサ抵抗は室温で 15Ω、転移点直上で 7Ωであった。これは抵抗率にすると 11.5Ωcm および 5.8Ωcm に対応し、バルク単結晶等で確認されている値と合致する。これにより c 軸配向微小メサ構造が実現していることがわかった。得られたメサの c 軸方向の電流電圧特性は抵抗短絡型ジョセフソン接合特性に近く、トンネル型特性は見られなかった。また、マイクロ波照射によるシャピロステップも観察させず、メサ部分

に a 軸方位の微結晶が存在しそれがマイクロショートとなっていることが示唆された。微小メサを用いて測定した臨界電流は 10^4A/cm^2 であり、以上のことを裏付けている。$La_{1.9}Sr_{0.1}CuO_4$ 微小メサ構造が達成されたので、今後はマイクロショートの影響を受けないトンネル分光特性測定法を考案する必要がある。

４．ビスマス系高温超伝導体固有ジョセフソン接合を用いた短パルストンネル分光

高温超伝導体 $Bi_2Sr_2CaCu_2O_{8+\delta}$ のイントリンシックジョセフソン接合を利用した短パルス固有トンネル分光により超伝導ギャップおよび擬ギャップの大きさを評価した。超伝導ギャップの値はこれまで走査型トンネル分光で評価されたものとほぼ等しいが、キャリア不足ドープ領域においてはこれまでよりも小さい値が得られた。また擬ギャップは最適ドープ領域および過剰ドープ領域では超伝導ギャップとほぼ同じであるが、不足ドープ領域では擬ギャップがはるかに大きく二つのギャップは異なるものであることがわかった。特に不足ドープ領域では超伝導ギャップと擬ギャップが異なる位置に観察され、擬ギャップが超伝導ギャップと同じ性質で T_c 以上で連続に超伝導ギャップから擬ギャップに連続に移行するものではないということを示している。これまで、高温超伝導を説明するモデルが種々提案されてきており、その中には超伝導電子対は高温から形成されており、T_c において電子対の位相がコヒーレントになって超伝導が出現するというモデルがある。このモデルでは超伝導ギャップと擬ギャップが T_c で連続に移行するとされていた。しかし、今回の実験ではこの二つのギャップが異なるものであることが示された。このことは、超伝導電子対が超伝導転移温度よりも高い、擬ギャップの形成される温度からすでに形成されているのではないことを意味している。したがって、本実験結果はこれらの理論モデルを否定するものと理解することができる。以上のように、固有ジョセフソン接合を用いた層間トンネル分光により、高温超伝導の発現機構に関して以上の知見が得られたことは極めて重要なことであり、今後の研究展開の指針を明示するものとして重要な意義を有するものと考えられる。

５．短パルス固有トンネル分光法のエネルギー測定領域の拡大

パルス幅 1μs の短パルス電流を微小メサ試料に流す場合、実際には任意波形発生装置から電圧パルスが供給されるためにパルス立ち上がり時に瞬間的に大

電流が流れる。測定では数 10mA の電流が流れることになり、これが電界 migration や電界降伏を誘起して試料破壊をもたらす。実際、これまで電流電圧特性を 1 接合当たり 100mV を越えると試料破壊が頻発し、ほとんどの試料が破壊するという状態であった。1 接合当たり 100mV では超伝導ギャップは観察できても高温超伝導の機構解明に重要な擬ギャップを十分観察することができない。特に、不足ドープ領域においては擬ギャップが 100mV を越える場合があり測定できない。そこでこの問題を解決するため、過渡電流を抑制することを目的として、正弦波と組み合わせ連結パルス波形を用いるトンネル分光法を考案した。実際にこの改良短パルス固有トンネル分光法では電界降伏による試料破壊が全く見られず、最高電圧は 1 接合当たり 200mV 以上まで確実になり、試料によっては 300mV に達した場合もあることが明らかになり、連結波形短パルスが極めて有効であることを明らかにした。

改良短パルス固有トンネル分光により、$Bi_2Sr_2CaCu_2O_{8+\delta}$ のトンネルスペクトルにおいて、超伝導ギャップよりも高い位置にもうひとつのピークが存在することが明らかになった。このピークはこれまで角度分解光電子分光などで観察されていた dip and hump 構造と解釈することも可能であるが、擬ギャップとの関係も無視できない。この新しい構造は不足ドープ領域で顕著になる傾向を有することから、前記の Tc 以下における擬ギャップと超伝導ギャップが共存する状態とも考えられる可能性が大きく、今後その可能性を精査する必要がある。このように、短パルス固有トンネル分光法の改良による分光エネルギー範囲が拡大したことにより、擬ギャップに関する詳細な実験が可能になり、その起因を含めて今後重要な知見が明らかになることが期待される。

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Interlayer Tunneling Spectroscopy for Slightly Overdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$

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We have measured the interlayer tunneling characteristics of slightly overdoped $Bi_2Si_2CaCu_2O_{8+\delta}$ 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]

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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 $Bi_2Sr_2CaCu_2O_{8+\delta}$ crystal by using fine processing technology. We employed these mesas to measure the tunneling characteristics of slightly overdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ 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. $Bi_2Sr_2CaCu_2O_{8+\delta}$ 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 μm × 20 μm. 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 Ω 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 $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals with a known oxygen content [6]. The mesas were, therefore, slightly overdoped.

Previously used $Bi_2Sr_2CaCu_2O_{8+\delta}$ 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 μs 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.

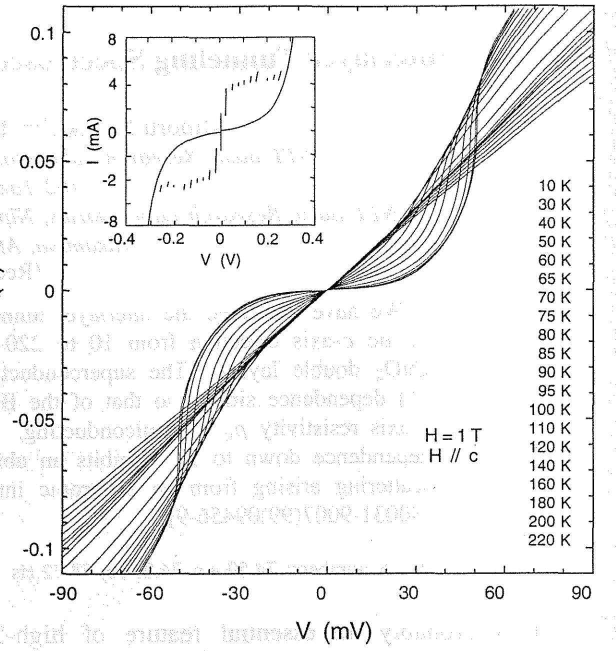

FIG. 1. The I-V characteristics of a $Bi_2Sr_2CaCu_2O_{8+\delta}$ mesa containing 11 Josephson junctions measured with 1 μs current pulses at temperatures from 10 to 220 K. A magnetic field parallel to the c axis was applied to suppress the Josephson current. The inset is an oscilloscope trace of the 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 $Bi_2Sr_2CaCu_2O_{8+\delta}$. 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 $Bi_2Sr_2CaCu_2O_{8+\delta}$ 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

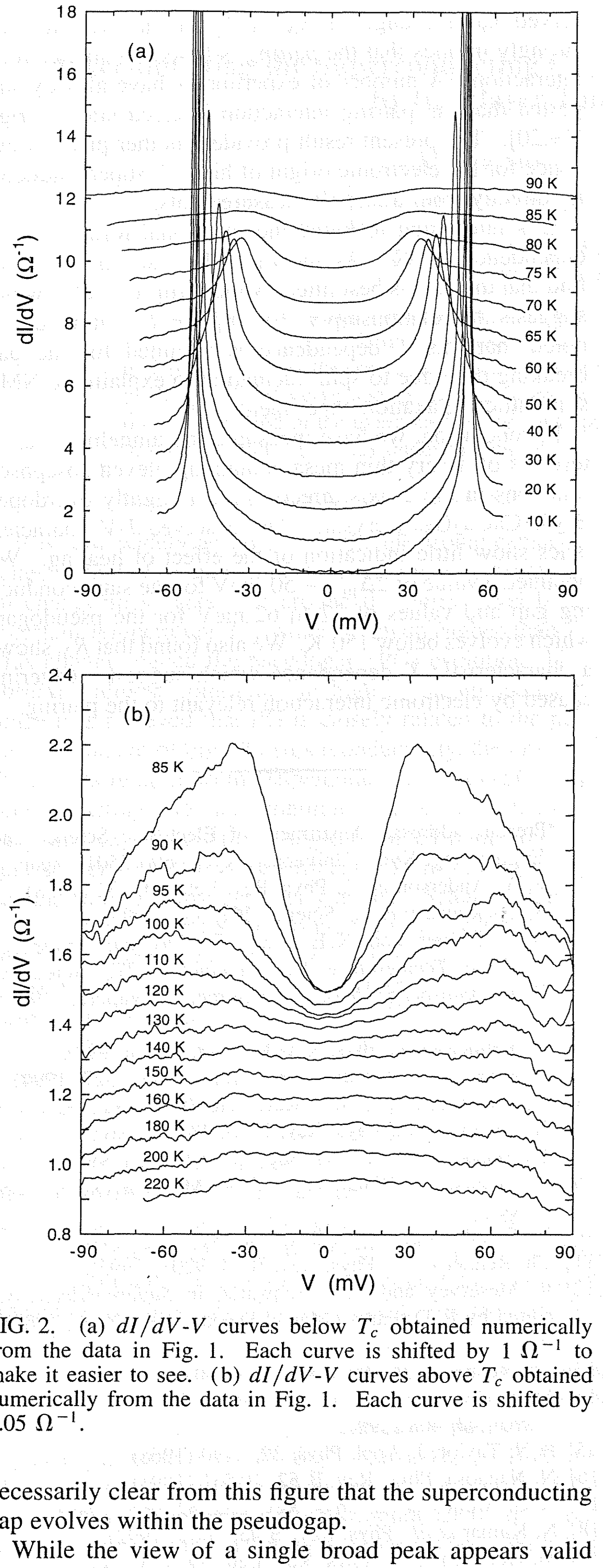

FIG. 2. (a) dI/dV-V curves below T_c obtained numerically from the data in Fig. 1. Each curve is shifted by 1 Ω^{-1} to make it easier to see. (b) dI/dV-V curves above T_c obtained numerically from the data in Fig. 1. Each curve is shifted by 0.05 Ω^{-1}.

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 $Bi_2Sr_2CaCu_2O_{8+\delta}$ 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

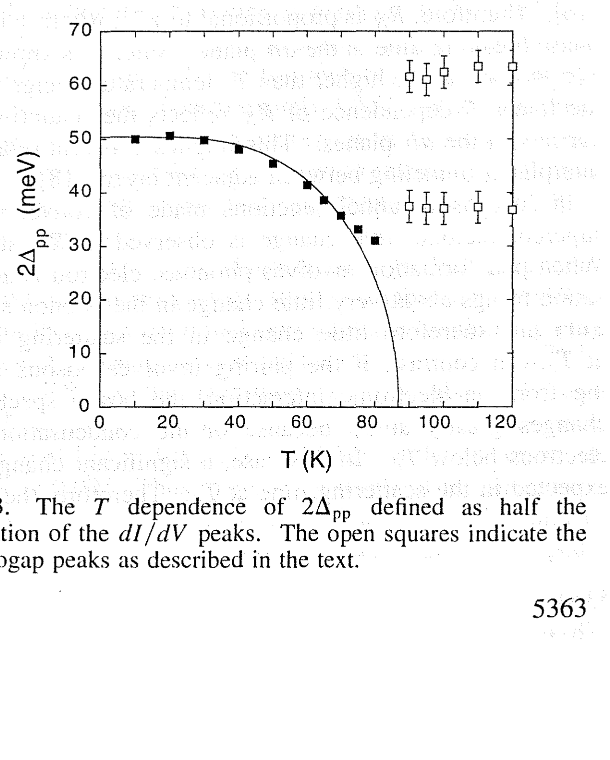

FIG. 3. The T dependence of $2\Delta_{pp}$ defined as half the separation of the dI/dV peaks. The open squares indicate the pseudogap peaks as described in the text.

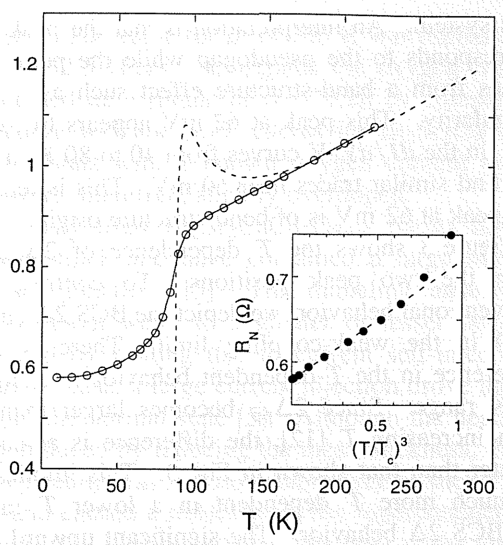

FIG. 4. The T dependence of R_N. The dashed line shows the T dependence of R_c/N. The inset shows the relationship R_N vs T/T_c. The solid line in the main panel and the dashed line in the inset are guides to the eyes.

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 observed 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 $Bi_2Sr_2CaCu_2O_{8+\delta}$ crystal. The observed I-V characteristics show little indication of the effect of heating. We obtained a value of $2\Delta_{pp} = 50$ 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.

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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 $Bi_2Sr_2CaCu_2O_{8+\delta}$ 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_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 $Bi_2Sr_2CaCu_2O_{8+\delta}$ (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 consequence 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 μm 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 μs wide current pulses at a duty of 0.05%. The influence of heating due to self-injection of current is less than 3% on the voltage scale for 20 μm 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 μm 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 $r = \rho_c^{\max}/\rho_c^{300\ \mathrm{K}}$, where $\rho_c^{\max}$ is the maximum ρ_c just above T_c and $\rho_c^{300\ \mathrm{K}}$ 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),

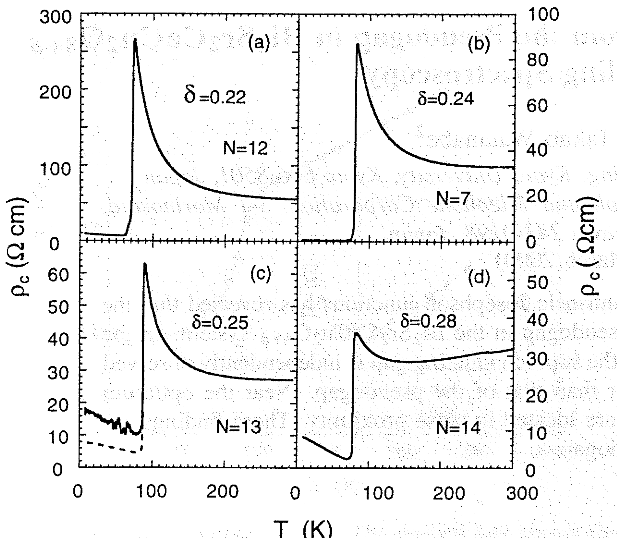

FIG. 1. Temperature dependence of the c-axis resistivity calculated from the stack resistance R_c for four specimens having different carrier doping levels: (a) $\delta = 0.22$ (underdoped), (b) $\delta = 0.24$, (c) $\delta = 0.25$ (near optimum), (d) $\delta = 0.28$ (overdoped). The dashed line in (c)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

respectively, as indicated in Fig. 1 together with a value for T_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

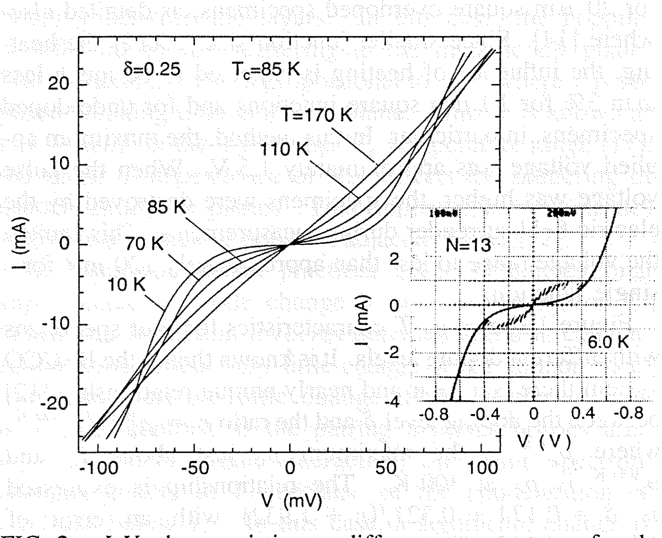

FIG. 2. I-V characteristics at different temperatures for the specimens in Fig. 1(c). The inset displays the oscilloscope I-V characteristics measured at 6 K, in which the lower branch is used for the tunneling spectroscopy.

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_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-V curve near T_c (thick line). The estimates of the peak position are 150 ± 20 mV, 130 ± 20 mV, 80 ± 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

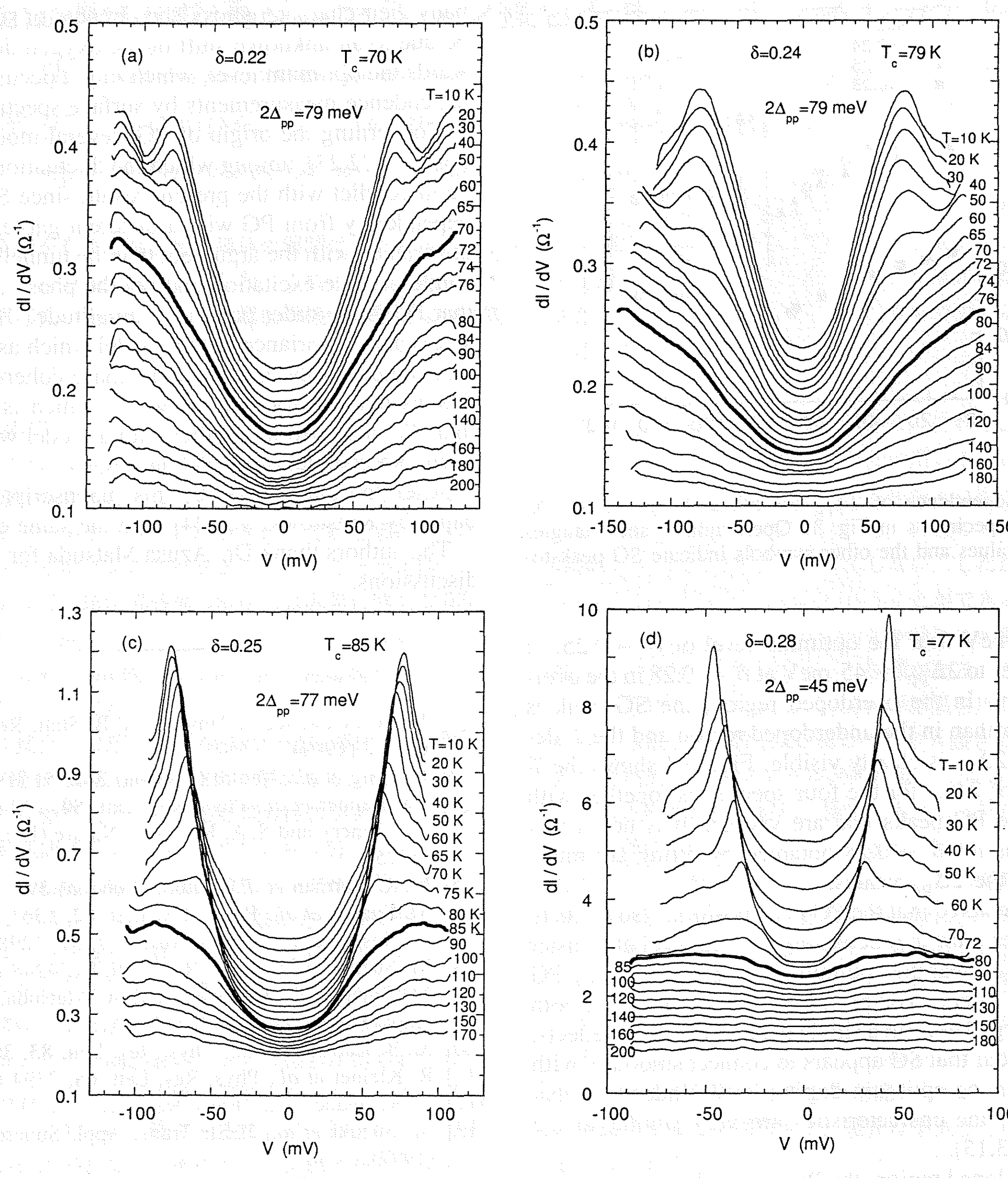

FIG. 3. dI/dV-V curves at various temperatures for the specimens in Fig. 1. Each curve is shifted vertically for an appropriate value. The thick lines indicate the curve very close to T_c. The normal tunneling resistance for the specimen in (d) exhibited a characteristic T dependence very similar to 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 optimum 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 $YBa_2Cu_3O_{7-x}$ [17–19] and $La_{2-x}Sr_xCuO_4$ [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

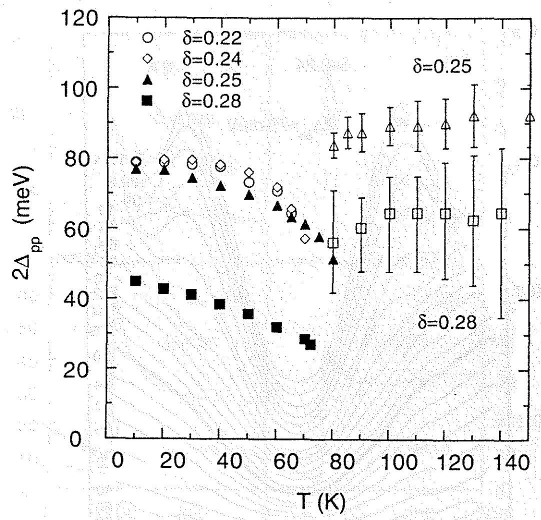

FIG. 4. T dependence of $2\Delta_{pp}$ for SG and, partly, for PG for the same specimens in Fig. 3. Open squares and triangles indicate PG values and the other symbols indicate SG peak-to-peak values.

$2\Delta_{pp} = 77$ 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 accompany 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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