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Electron-tunneling studies on the superconducting $(La_{1-x}Sr_x)_2CuO_4$ and $YBa_2Cu_3O_y$ systems

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We report the results of tunneling measurements on $(La_{1-x}Sr_x)_2CuO_4$ and $RBa_2Cu_3O_y$ (R=Y,Er,Gd) at several x and y values having various T_c's. The ratios $2\Delta/kT_c$ are 4.5–4.9 for $(La_{1-x}Sr_x)_2CuO_4$ and 3.3–4.2 for $RBa_2Cu_3O_y$. The shapes of the tunneling density of states, anisotropies of the gaps, and temperature dependences of the gaps are also discussed.

Electron-tunneling studies on high-T_c oxide superconductors have been performed to understand the nature of these materials. This technique offers one of the most direct ways to elucidate the mechanism by measuring the superconducting energy gap and excitation spectrum reflected in the electronic density of states. In the early stages of high-T_c tunneling studies, many unexpected characteristics have been reported: for example, extremely large gaps, multiple-gap structures, strong asymmetric tunneling conductances with respect to the zero bias voltage, etc. These phenomena have indicated nonintrinsic tunneling characteristics for the cuprate superconductors. At present there is still no common understanding of the superconducting tunneling density of states. Besides this problem, it is well known that $(La_{1-x}Sr_x)_2CuO_4$ and $YBa_2Cu_3O_y$ show the superconducting-antiferromagnetic transition just after the disappearance of superconductivity at about $x=0.02$ for $(La_{1-x}Sr_x)_2CuO_4$ and $y=6.5$ for $YBa_2Cu_3O_y$. Are the superconducting energy gaps substantially the same for the different compositions and/or different T_c?

In this paper, we report conclusive results of electron-tunneling measurements of the energy gaps in $(La_{1-x}Sr_x)_2CuO_4$ with $x=0.025-0.3$ and $YBa_2Cu_3O_y$ with $y=7$ and 6.6–6.7, especially clarifying the T_c dependences of energy gaps for both systems. We have analyzed the observed tunneling density of states using the broadened BCS density of states.[1]

Samples were prepared as follows. Mixtures of starting materials were pressed into pellets and synthesized by the solid-state reactions. A series of $(La_{1-x}Sr_x)_2CuO_4$ with $x=0.025$, 0.035, 0.045, and 0.30 were fired at 1050–1100 °C for 3–7 d under flowing oxygen gas and furnace cooled to room temperature. $RBa_2Cu_3O_y$ (R=y, Gd, Er) (T_c=90-K phase) were fired at 950 °C for Y and Er, 1050 °C for Gd under flowing oxygen gas for 1–2 d. They were annealed again at 600 °C in oxygen gas for 2–3 d and finally furnace cooled to room temperature. The samples of the T_c=50-K phase of $YBa_2Cu_3O_y$ were prepared by a gettered annealing technique.[2] The oxygen contents y of this phase were determined by powder x-ray diffraction and weight change before and after the annealing procedure. T_c's versus y coincide with those in Ref. 2. The superconducting transition temperature T_c was determined by the dc four-probe method, Meissner measurements, and also by the temperature dependence of the zero-bias conductance of the tunnel junction. $RBa_2Cu_3O_y$ (T_c=90-K phase) have shown a resistive transition width of within 1–2 K for the dc four-probe measurement. Tunnel measurements were done by the point contact junction with Al or Pt electrode. The typical junction area was about 0.1 mm^2. The native surface insulating layer of the sample has often formed a tunnel barrier, although its characteristics are unknown. Measurements of the dynamic conductances $dI/dV(V)$, where I and V are the tunnel current and dc bias voltage, respectively, were the standard ac-modulation method.

Figure 1 shows the raw data of $dI/dV(V)$ curves obtained from the $(La_{1-x}Sr_x)_2CuO_4$-Pt tunnel junctions for the Sr concentrations $x=0.025$, 0.035, and 0.045. The $dI/dV(V)$ curve for $x=0.075$ has already been reported.[3] The resistance of the tunnel junction ranged within 20–1 kΩ. The superconducting tunnel conductance at zero bias $[=(dI/dV)_s(0\text{ mV})]$ at low temperature reduces only 15–20 % of the normal-state zero-bias conductance $[=(dI/dV)_n(0\text{ mV})]$ interpolated from a

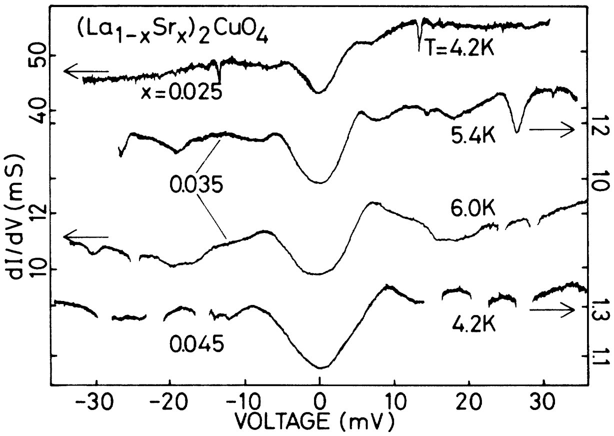

FIG. 1. The $dI/dV(V)$ curves from various Sr concentrations x in $(La_{1-x}Sr_x)_2CuO_4$ at low temperatures. The $dI/dV(V)$ curve for $x=0.075$ has been reported separately (Ref. 3).

higher-bias region, at any measurement. A large leakage current may be due to tunneling into normal material. These fractions do not change drastically with x in spite of the superconducting volume fractions change with concentrations, namely more than 70% for $x=0.045$ to about 30% for $x=0.025$. As shown in Fig. 1, there is no short circuit in the tunnel junctions and the $dI/dV(V)$ curves are less asymmetric. Structures outside the gaps are seen in every curve. Most of the structures at higher-bias voltages are not intrinsic. However, we observed the reproducible structures at about 15–20 mV, for example, $dI/dV(V)$ curves at $x=0.035$ in Fig. 1. These structures were also observed by Bulaevskii *et al.*[4] and reported as phonon density of states.[5]

Since the observed $dI/dV(V)$ curves are broadened, the peak-to-peak value of the $dI/dV(V)$ curve does not correspond to the gap value in the BCS expression so that, to determine the energy gap value 2Δ, we have employed the broadened BCS density of states

$$D(E,\Gamma)=\mathrm{Re}\{(E-i\Gamma)/[(E-i\Gamma)^2-\Delta^2]^{1/2}\}$$

(Ref. 1), where Γ is a broadening parameter, and fitted to the selected experimental data of $dI/dV(V)$ which were symmetric with respect to zero bias and flat against the bias voltage since the gap edge with a strongly bias-dependent conductance was often largely broadened. It should be noted that there are mainly two types of destruction of superconductivity: reduction in amplitude of the order parameter Δ and breaking of phase coherence which is related to Γ. Breaking of phase coherence makes the superconducting density of states broadened, i.e., it yields finite quasiparticle states within the gap region even at $T=0$ K.[6] In the fitting procedures we have subtracted a large fraction of the tunneling conductance which can be attributed to tunneling into the normal material rather than an intrinsic mechanism and attained correspondence between the data and the theoretical curve. One of the fitting results in $(La_{1-x}Sr_x)_2CuO_4$ is shown as curve A in Fig. 5 for $x=0.035$ at $T=7.9$ K. The best-fitted values in this case are $2\Delta=9.8$ meV and $\Gamma=1.3$ meV. This agrees with the voltage difference where $(dI/dV)_s(V)$ and $(dI/dV)_n(V)$ cross each other. The other Sr concentrations, $x=0.025$ and 0.045, in Fig. 1 have the gap values $2\Delta=5$ and 12 meV, respectively. The smaller gap at $x=0.035$ in Fig. 1 will be discussed later.

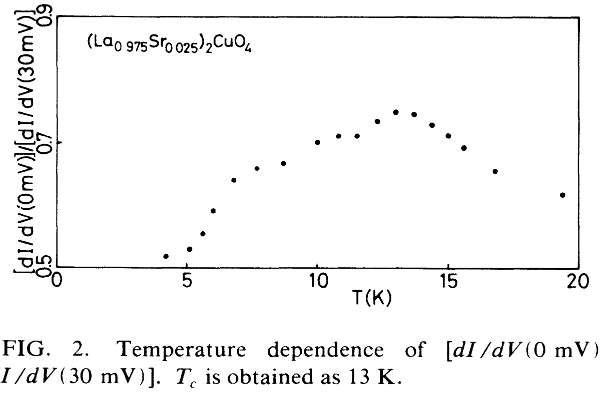

FIG. 2. Temperature dependence of $[dI/dV(0\text{ mV})]/[dI/dV(30\text{ mV})]$. T_c is obtained as 13 K.

To obtain the ratio $2\Delta/kT_c$, the T_c's were determined from the temperature where $dI/dV(0\text{ mV})$ begins to reduce. Figure 2 shows a temperature dependence of $[dI/dV(0\text{ mV})]/[dI/dV(30\text{ mV})]$ of $(La_{1-x}Sr_x)_2CuO_4$ ($x=0.025$). In this sample, the decrease of $dI/dV(0\text{ mV})$, i.e., the opening of the energy gap due to the onset of the superconductivity at the tunnel junction, begins at $T=13$ K, whereas the onset of T_c from the resistance measurement is 20 K and end point 10 K. By employing $2\Delta=5$ meV, $2\Delta/kT_c$ is 4.5. The T_c's for $x=0.035$ and 0.045 are 23–25 and 30 K, respectively, so that the ratios $2\Delta/kT_c$ are 4.5–4.9. These values almost coincide with that of $x=0.075$ which has the maximum T_c (= 35 K) in the $(La_{1-x}Sr_x)_2CuO_4$ system.[3,7,8] Therefore, we conclude that the ratio $2\Delta/kT_c$ remains constant although T_c decreases by a factor of 3 by changing the Sr concentration x. We have found the double-gap values as shown at $x=0.035$ in Fig. 1. These gap values are 9.8

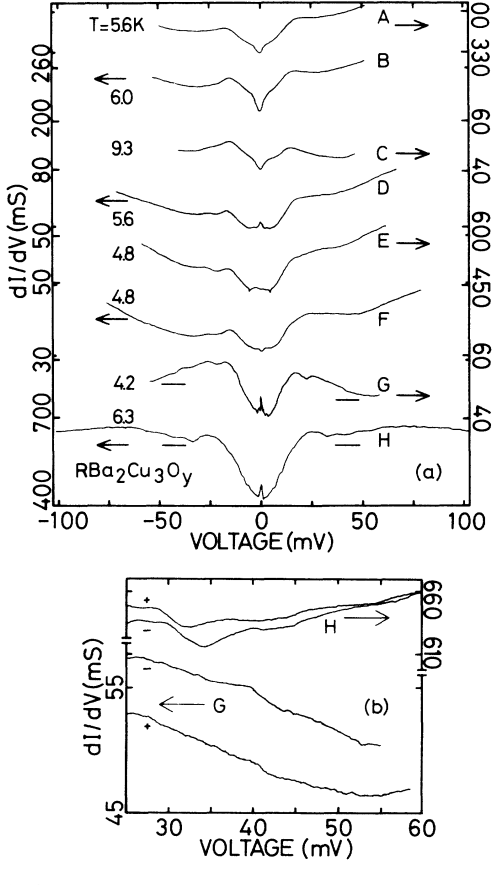

FIG. 3. (a) Assembly of the raw data of the $dI/dV(V)$ curves for $YBa_2Cu_3O_y$ (A,B,D,G,H), $ErBa_2Cu_3O_y$ (C,E), and $GdBa_2Cu_3O_y$ (F) at low temperatures. Samples used in the present experiments are the $T_c=90$-K phase. A variety of curves can be systematically classified. Double-gap structures are apparently observed in the curves D, E, F, and G. Bars at about ±40 mV under the curves G and H show the bias positions of the characteristic structures. (b) The structures in the $dI/dV(V)$ curves of $YBa_2Cu_3O_y$. The upper two curves are the same as H and the lower two are G in Fig. 3(a). The + and − signs show the bias polarity.

and 8.4 meV, and the ratio of gaps about 1.2 is the same order as the $YBa_2Cu_3O_y$ system, as will be described later.

Figure 3(a) shows the $dI/dV(V)$ curves of $RBa_2Cu_3O_y$ (R=Y, Sr, Gd). The superconducting zero-bias conductances are about 80–85 % of normal zero-bias conductance interpolated from higher-bias regions, and this fraction was nearly the same as any sample of $RBa_2Cu_3O_y$ and $(La_{1-x}Sr_x)_2CuO_4$. A set of curves A, B, D, G, and H [we abbreviate (A,B,D,G,H)], (C,E), and F correspond to R=Y, Er, and Gd, respectively. The tunnel resistances at the higher voltages were 1–20 Ω. As shown in Fig. 3(a), $dI/dV(V)$ curves are classified into four types in shape, namely, (A,B,C), (D,E,F), G, and H. Double peaks exist in $dI/dV(V)$ curves (D,E,F,G) and the peaks of the positive bias in (D,E,F) are smeared. Note that the bias voltage refers to the counter electrode. The inner and outer peaks at the bias positions of about ±16 and ±26 meV in (D,E,F,G) correspond to those of (A,B,C) and H, respectively. The set of the single peak as shown in (A,B,C) and H may correspond to each part of the two peaks which appear in (D,E,F,G). The shapes and the bias positions of peaks in $dI/dV(V)$ are well reproducible at any sample. The ratio of the peak-to-peak values in $dI/dV(V)$ for H (outer peak) to (A,B,C) (inner peak) is 1.6. This value coincides with the gap anistropy with respect to the CuO_2 plane reported by Tsai *et al.*[9] The gaps from the multiple-band structure as predcited by Kresin[10] and by Schopohl and Scharnberg[11] can also be an origin. The conductance dip in the gap around zero bias with the width of about 8–11 meV was consistently observed in the smaller-gap structure as shown in curves A, B, and C. A similar dip was also confirmed by Fournel *et al.*[12] and Gurvitch *et al.*[13] using the single crystals. One of the possible origins is that one observes the tunneling density of states of the surface layer which is superconducting due to the proximity effect backed by the intrinsic superconducting bulk sample.[14] gap anisotropy is also considered as the origin of this structure.[13] This dip begins to develop below 25–30 K (Refs. 13 and 14) suggesting an occurrence of a phase transition at this temperature. The ratio of 8–11 meV to 25–30 K is 3.1–5.1. Curves G and H in Fig. 3(a) show a gentle ($\simeq 1-2\,\%$) and sudden degradation of $dI/dV(V)$ with an increase of bias voltage at about $V=\pm 40-45$ mV ($eV-\Delta \simeq 25-30$ meV). This bias position is well reproducible and the details are shown in Fig. 3(b). The upper and lower two curves in Fig. 3(b) are the same as H and G in Fig. 3(a), respectively. This structure seems to appear when $dI/dV(V)$ is nearly independent of the bias voltage. The same structure has been reported by some other authors. Lee *et al.*[15] observed the structures at V=20 and 40 mV for several planar thin-film $YBa_2Cu_3O_y$ tunnel junctions. Gurvitch *et al.*[13] also reported a structure at $V=\pm 42-50$ mV in the single-crystal junction. This reproducible structure can be due to the renormalized superconducting density of states due to a strong-coupling effect with the excitation spectra which is speculated from the structure similar to the Eiliashberg interaction which occurs at the characteristic frequency ω_c and appears as a correction of $1.5(\Delta/\omega_c)^2$ to the normal tunnel conductance.

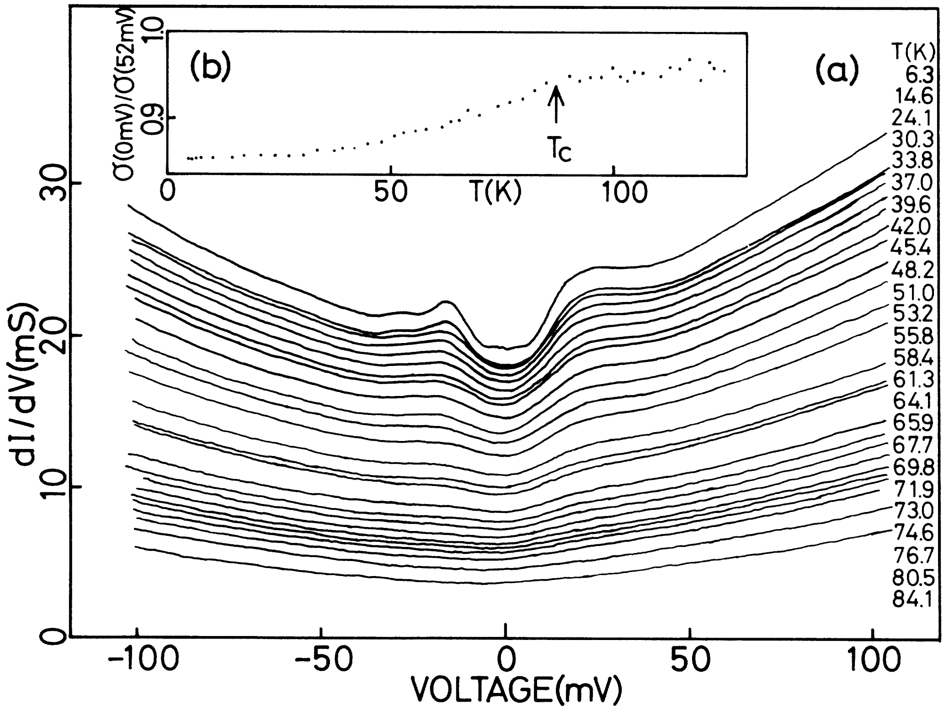

FIG. 4. (a) Temperature dependence of $dI/dV(V)$ curves of $GdBa_2Cu_3O_y$ (T_c=90-K phase). The bias voltage refers to the aluminum counter electrode. (b) Temperature dependence of $[dI/dV(0\text{ mV})]/[dI/dV(52\text{ mV})]=\sigma(0\text{ mV})/\sigma(52\text{ mV})$. T_c is assigned unambiguously as 87 K.

Figure 4(a) shows the temperature variation of $dI/dV(V)$ curves of $GdBa_2Cu_3O_y$ up to near T_c. In this figure, the double peaks in $dI/dV(V)$ at the same bias positions as in Fig. 3(a) also appear. Tunnel conductance in this case monotonically decreases with the increase of temperature. T_c was unambiguously determined to be 87 K from the plot of the temperature dependence of

$$[dI/dV(0\ \mathrm{mV})]/[dI/dV(52\ \mathrm{mV})] = \sigma(0\ \mathrm{mV})/\sigma(52\ \mathrm{mV})$$

as shown in Fig. 4(b). This is about 7 K lower than the zero-resistance temperature.

Figure 5 (B,C,D,E,F,G,H,I) shows the fitting results for several $dI/dV(V)$ curves of $RBa_2Cu_3O_y$ using $D(E,\Gamma)$. The data used for the fitting are normalized by $(dI/dV)_n(V)$ interpolated from the higher-bias regions. Curves (B,C) are the $T_c=90$-K phases of $YBa_2Cu_3O_y$ (curve D is the $T_c=50$-K phase and will be discussed later). Curves (E,G,H,I) are $GdBa_2Cu_3O_y$ and F is $ErBa_2Cu_3O_y$. As already described in the fitting procedure for $(La_{1-x}Sr_x)_2CuO_4$, we regard the large fraction of zero-bias conductance as tunneling into normal material so that in the fitting we have subtracted a certain fraction from measured conductance. In $RBa_2Cu_3O_y$ at $T=4$ K, the calculated normalized zero-bias conductance due to the gap region broadening using the relation

$$dI/dV(0\ \mathrm{mV},\ T=0\ \mathrm{K})=D(0,\Gamma)=\Gamma/(\Delta^2+\Gamma^2)^{1/2}$$

is 0.3–0.4, which is about 10% of the total normal zero-bias conductance interpolated from the higher-bias region. The remaining conductance, about 70%, is speculated as due to tunneling into normal material. 2Δ obtained with the fitting of the $T_c=90$-K phase samples has two distinct gap values as shown in (B,E,F) and (C,G). The larger gap is $2\Delta=30$–31 meV with $\Gamma=4$–5.5 meV from (C,G,H) and the smaller gap is $2\Delta=24$–26 meV with $\Gamma=3$–5 meV from (B,E,F) so that the ratios $2\Delta/kT_c$ are 4–4.1 and 3.2–3.5 using T_c determined in similar manner as shown in Fig. 4(b). The ratio $2\Delta/kT_c$ less than 3.5 other than the gap anisotropy can be explained by the two-band model of Schopohl and Scharnberg[11] and Kresin[10] where it always less than 3.5 for the smaller gap in the system with two gaps. The ratio of two gaps is about 1.3 (the ratio taken as the peak-to-peak value is 1.6), and this is almost the same as that of $(La_{1-x}Sr_x)_2CuO_4$, as already mentioned, but considerably smaller than that of the Bi-Sr-Ca-Cu-O systems.[16] The meaning of Γ in this case is not obvious, but reproducible for the different junctions. The large ratio of $\Gamma/\Delta=0.3$–0.5 at low temperatures is possibly due to the short coherence length of this system and/or surface inhomogeneity of tunnel junctions. A modified tunneling density of states from the BCS one due to the breaking of phase coherence is reflected not in Δ but in Γ.[18] Therefore, it is expected that the fitting procedures using Δ and Γ uniquely determine the energy gap 2Δ.

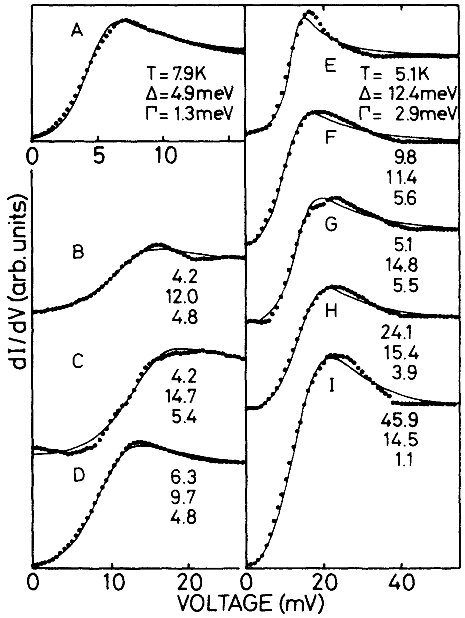

FIG. 5. The results of fitting. Solid circles and solid lines are the experimental data and the theoretical curves given by $D(E,\Gamma)$ with thermal broadening, respectively. Curve A corresponds to $(La_{0.965}Sr_{0.035})_2CuO_4$, (B,C) to $YBa_2Cu_3O_y$ ($T_c=90$-K phase), D to $YBa_2Cu_3O_y$ ($T_c=50$-K phase), (E,G,H,I) to $GdBa_2Cu_3O_y$, and F to $ErBa_2Cu_3O_y$.

The temperature dependences of the two energy gaps of $GdBa_2Cu_3O_y$ are shown in Fig. 6(a). These gap values are extracted from the same procedures as in Fig. 5. As indicated in Fig. 6(a), the two gaps well obey the scaled BCS curves and seem to decrease toward the same T_c. Therefore, these two gaps are not attributed to the gaps of different superconducting phases but to the gap anisotropy. The gap ratio seems to temperature independent at least up to 60 K ($T/T_c=0.7$).

For the tunnel junction of superconductor-insulator-normal (SIN) configuration with no short circuits, the peaks in $dI/dV(V)$ with increasing temperatures are only broadened and the peak position does not shift to a lower bias.[6] On the contrary, a tunnel junction with low-resistance contact containing microscopic short circuits, i.e., narrow conducting channel in the insulating layer, makes it possible to observe the singularity of the tunnel current in the superconducting state[19] and the bias position of this singularity obeys the BCS temperature dependence.[20] Many reports have tracked this characteristic of the temperature variations.[4,21–24]. Figure 6(b) shows the temperature variation from this type of junction for $(La_{1-x}Sr_x)_2CuO_4$ and $YBa_2Cu_3O_y$ obtained from our data. The BCS temperature variations of superconducting order parameters are obviously seen. These results confirm that the temperature dependence of superconducting energy gaps are well described by the scaled BCS curves.

$YBa_2Cu_3O_y$ has the phase, of which T_c is reduced to about 50 K at $y=6.6$–6.7.[2] Such a reduction is the consequence of the decrease of the effective hole-carrier concentrations. We measured the gaps at $y=6.6$–6.7 to clarify the energy gaps for the change of y. Figure 7 shows the $dI/dV(V)$ curves of $YBa_2Cu_3O_y$ with

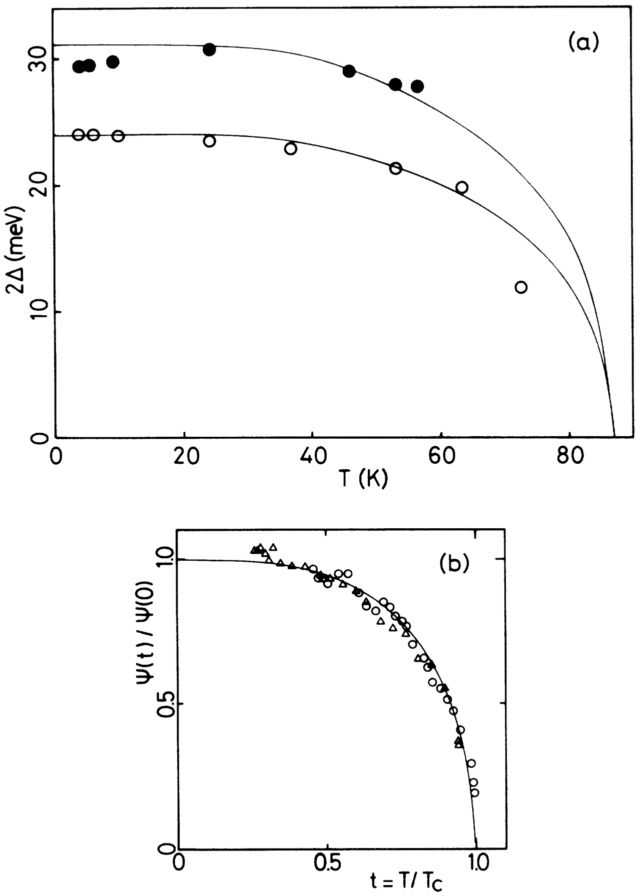

FIG. 6. (a) Temperature dependences of two energy gaps for $R\mathrm{Ba_2Cu_3O}_y$ obtained by the use of $D(E,\Gamma)$. The solid lines are the scaled BCS curves. (b) Temperature dependences of the peak-to-peak values in $dI/dV(V)$ curves $[=\Psi(t)]$ normalized by their zero-temperature values. Open triangles and open circles correspond to $(\mathrm{La}_{1-x}\mathrm{Sr}_x)_2\mathrm{CuO}_4$ and $\mathrm{YBa_2Cu_3O}_y$, respectively. Those show BCS temperature dependences. Details are described in the text.

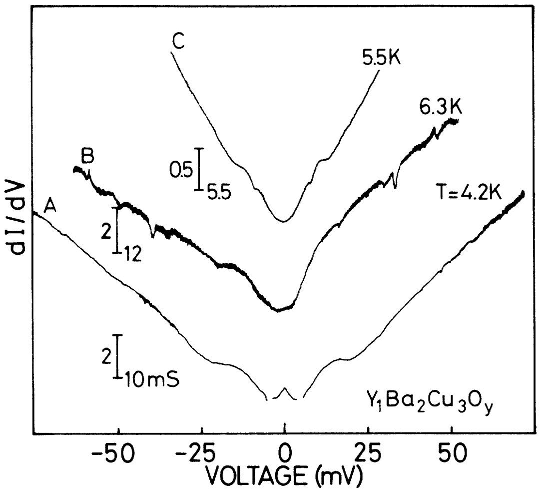

FIG. 7. The $dI/dV(V)$ curves for $\mathrm{YBa_2Cu_3O}_y$ ($T_c=50$-K phase) Curves A, B, and C are obtained from different tunnel junctions.

$y=6.6-6.7$. The temperature dependence of the resistivity ρ of these samples just above the transition temperature shows metallic behavior, i.e., $d\rho/dT>0$. The typical tunnel resistances are higher than that of the $T_c=90$-K phase samples. In Fig. 7, the $dI/dV(V)$ curves named A, B, and C are taken from different batches of samples. The $[(dI/dV)_s(0\ \mathrm{mV})]/[(dI/dV)_n(0\ \mathrm{mV})]$ are about 0.8. The strong linear bias dependences of $dI/dV(V)$ may not be intrinsic because, as seen in Figs. 1, 3, and 4(a), the weak bias dependence of $dI/dV(V)$ were also observed. It may depend on the quality of the tunnel barrier. One of the fitting results for the $T_c=50$-K phase is shown to curve D ($T_c=55$ K) in Fig. 5. The gap values of curves in Fig. 7 are $2\Delta=18-19$ meV so that the ratio $2\Delta/kT_c$ is 4.1–4.7 employing the T_c of 47–55 K. This value is within the range of the $T_c=90$-K phase sample. We therefore confirm that $2\Delta/kT_c$ does not change when T_c reduces to 50 K towards the superconducting-antiferromagnetic transition region. The situation is same as that of $(\mathrm{La}_{1-x}\mathrm{Sr}_x)_2\mathrm{CuO}_4$ with decreasing x. However, Rajam *et al.*[25] have concluded that $2\Delta/kT_c$ in $\mathrm{YBa_2Cu}_{3-x}\mathrm{Zn}_x\mathrm{O}_y$ monotonically decreases with an increase of the zinc concentrations x (decrease of T_c). This result is in contrast with ours and can be explained by the partial substitution of the zinc ions at the copper-ion sites in $\mathrm{CuO_2}$ planes increasing randomness and directly violating the interaction (e.g., Cu spin interaction) as an operative mechanism of the superconducting and leading to the depression of the strong-coupling nature of them. On the other hand, a decrease of T_c by only decreasing carrier holes on $\mathrm{CuO_2}$ planes, as described in this paper makes no change in the strong-coupling nature of the superconductivity.

In our analysis, we introduced the broadened BCS density of states $D(E,\Gamma)$ to determine the superconducting energy gap. The observed tunneling density of states can be expressed by $D(E,\Gamma)$. Therefore, we conclude that the gap values extracted by $D(E,\Gamma)$ are reliable. Many reports on the tunneling measurements have revealed the ratios $2\Delta/kT_c=4-7$ for both $(\mathrm{La}_{1-x}\mathrm{Sr}_x)_2\mathrm{CuO}_4$ and $\mathrm{YBa_2Cu_3O}_y$. One of the reasons for the different $2\Delta/kT_c$ among the several reports is related to the methods for gap determinations. This can be qualitatively discussed as follows. If we take 2Δ as peak-to-peak value in $dI/dV(V)$, it is about 1.3–1.8 times larger than the fitting result, namely $2\Delta/kT_c=6.2-8.6$ for $(\mathrm{La}_{1-x}\mathrm{Sr}_x)_2\mathrm{CuO}_4$ and 5.5–7.6 for $\mathrm{YBa_2Cu_3O}_y$. Moreover, Deutscher and Muller[26] discussed the weakening of the gap at the sample surface in high-T_c superconductors that have a short coherence length and refer to the tunneling results underestimating the gap values. Another reason for the discrepancy of gap values among the reports is possibly due to the anisotropy of gaps or the gaps of multiband structure as already discussed in this article.

The origin of the periodic structure in the high-T_c superconducting tunneling current was discussed by Barner and Ruggiero[27] and Bentum *et al.*[28] They observed the tunneling characteristics for the systems consisting of small isolated metal particles embedded between two tunneling electrodes. These $I(V)$ characteristics that are stepwise refer to the Coulomb staircase. Another explanation is the enhancement of the density of states at energies that satisfy the resonance conditions of the quasiparticle excitations as discussed by Tomasch.[29]

We briefly mention the gaps obtained from other techniques complementary to the tunneling measurements. The nuclear magnetic relaxation experiment for $(La_{1-x}Sr_x)_2CuO_4$ results in $2\Delta/kT_c=7$,[30] whereas Schlesinger *et al.*[31] observed 2.5 from far-infrared (FIR) measurements along the c axis. As for $YBa_2Cu_3O_y$, the Andreev reflection measurement at the Ag-$YBa_2Cu_3O_y$ interface[32] shows the zero-momentum (s-wave) pairing and the BCS value of $2\Delta/kT_c(=3-4)$. It shows a clear decrease in point contact resistance at the bias position of $\pm10.5-14.5$ meV which corresponds to the gap energy. The ac susceptibility measurement[33] and the ultrasonic attenuation experiment[34] give $2\Delta/kT_c=5$ and 3.5, respectively. The ratio $2\Delta/kT_c$ for $YBa_2Cu_3O_y$ from several experimental techniques tend to be smaller than that of $(La_{1-x}Sr_x)_2CuO_4$. However, the FIR measurements by Schlesinger *et al.*[35] and Collins *et al.*[36] on $YBa_2Cu_3O_y$ single-crystal a-b plane reflectivity give $2\Delta/kT_c=8$, which is much larger than that obtained from other techniques. They also observed a smaller gap $2\Delta/kT_c=3$ along the c-axis reflection. This suggests the substantial gap anisotropy in this system. The consistent $2\Delta/kT_c$ value with the tunneling results was also observed by the FIR measurement.[37] The extremely large values of the absorption energy edge reported from the FIR measurements was possibly due to strong dispersion in the real part of the dielectric function or the changes in the frequency-dependence scattering rate so that this absorption cannot be attributed to the superconducting gaps.[38]

We discuss the carrier concentration dependence of the energy gap including excess hole region. The hole carriers are excessively doped in the CuO_2 planes for an increase of x in $(La_{1-x}Sr_x)_2CuO_4$ above $x=0.075$ and the partial substitution of Y by the Ca ion in $YBa_2Cu_3O_y$. We have previously observed the extremely large gap of 30–35 meV taken as peak-to-peak values in $dI/dV(V)$ and/or 24 meV obtained from $(dI/dV)_n=(dI/dV)_s$ on $La_{1.7}Sr_{0.3}CuO_4$ with a T_c of less than 4 K and the background conductance in this case was quite metallic (flat) at least up to ±100 mV. A natural explanation is the gap value of a higher-T_c phase. Fein *et al.*[39] also observed the extremely large gap for $(La_{1-x}Sr_x)_2CuO_4$ with $x=0.115$ and they conjectured that this was attributed to the broad transition temperature width. The gap value of excess hole concentrations $Y_{1-x}Ca_xBa_2Cu_3O_y$ was measured by us as $2\Delta=35$ meV; thus, $2\Delta/kT_c=4.8-5.4$ with $T_c=85-76$ K from the resistivity measurement.

We now try to calculate the T_c of $(La_{1-x}Sr_x)_2CuO_4$ from the $dI/dV(V)$ curve at $x=0.035$ and $T=6$ K in Fig. 1 using the Allen-Dynes T_c equation.[40] In this calculation, we assumed the coupling constant λ to be about 2 from the deviation about 4% from the background conductance curve at $V=15-20$ mV and $\langle\omega\rangle$ taken as the energy eV-$\Delta\simeq18-5$ meV $=$ 13 meV, where $\langle\omega\rangle$ is the characteristic average frequency.[40] T_c is then obtained as 22 K with the assumed effective Coulomb repulsion $\mu^*=0.1$. This is in agreement with the measured value of 23–25 K. For $YBa_2Cu_3O_y$, T_c is calculated as 22–35 K for $\lambda=1-1.5$ from a 2% deviation in $dI/dV(V)$ and $\langle\omega\rangle\simeq42-15$ meV$=27$ meV from Fig. 3(b) and assumed $\mu^*=0.1$, which is largely inconsistent with the measured T_c of 90 K; however, some correction of μ^* can realize the measured T_c.

Mitrovic *et al.*[41] empirically calculated the $2\Delta/kT_c$ of the strong-coupling superconductors from the Eliashberg equation as a function of $T_c/\omega_{\ln}$, where $\omega_{\ln}$ is the logarithmic average frequency given by Allen and Dynes.[40] Marsiglio *et al.*[42] discussed the limitation of the Eliashberg theory on high-T_c superconductors. We discuss using their relation

$$2\Delta/kT_c(T_c,\omega_{\ln})=3.5[1+12.5(T_c/\omega_{\ln})^2\ln(\omega_{\ln}/2T_c)]$$

(Ref. 41), assuming the s-wave pairing thus ignoring gap anisotropy. By employing $T_c=35$ K and $\omega_{\ln}=130$ K for $(La_{1-x}Sr_x)_xCuO_4$ at $x=0.075$, $2\Delta/kT_c$ is obtained as 5.5 and it coincides with the experimental results 4.7–5.2.[3,7] Our result of the x dependence of $2\Delta/kT_c$ and also the result by Fein *et al.*[39] suggest that $T_c/\omega_{\ln}$ remains constant even though T_c decreases. As for the $T_c=90$-K phase of $YBa_2Cu_3O_y$, it also becomes about 5.5 if we take $\omega_{\ln}=260$ K, which is different from our observed values 4–4.2. We conversely obtain $\omega_{\ln}=900$ K from measured $2\Delta/kT_c=4-4.2$. If the Eliashberg theory is still valid in $YBa_2Cu_3O_y$, this suggests that some kind of high-energy excitation mediates the electron pairing and leads to high-T_c superconductivity. Note that the effective exchange interaction of the Cu spins of the CuO_2 plane in the copper oxides is an order of 1000 K.

As described in this article our significant result is that the ratios $2\Delta/kT_c$ in both systems are independent of T_c, i.e., hole concentrations. On the contrary, tunneling studies of the zinc-doped cuprate as mentioned before,[25] and noncuprate materials $BaPb_{1-x}Bi_xO_3$ (Ref. 43) and $A15$ materials Nb_3X (X=Sn, Al, Ge (Refs. 44 and 45) show that $2\Delta/kT_c$ increases with increasing T_c, namely, $2\Delta/kT_c$ varies from 3.5 to 5.6 ($T_c=50-91$ K) with the Zn concentrations $x=0.1-0.0$ in $YBa_2Cu_{3-x}Zn_xO_y$, 2–3.8 (76.–10.3 K) with the Bi concentrations $x=0.16-0.22$ in $BaPB_{1-x}Bi_xO_3$), and 3.0–4.5 (9.4–16.4 K) in Nb_3X with the concentration of X approaching stoichiometry. The latter two results suggest the enhancement of the electron-phonon interaction when the concentrations approach the phase boundary. Valles, Dynes, and Garno[46] found a marked difference in behavior of $2\Delta/kT_c$ in their tunneling measurement as a function of film sheet resistance between superconducting ultrathin quench-condensed Sn and Pb films, where $2\Delta/kT_c$ in the Sn film decreases from 4.5 to the BCS value or less when the sheet resistance increases (T_c decreases as 6 to 2.5 K), on the contrary it remains unchanged (4.5) in the Pb film. Besides the tunneling, the

FIR study reveals that the ratio $2\Delta/kT_c$ of Fe-doped $YBa_2Cu_3O_y$ decreases with Fe concentration[47] which is consistent with the Abrikosov-Gor'kov theory.[48] The fact that the ratio $2\Delta/kT_c$ remains constant or varies with T_c is the evidence that the different mechanism works on the decrease of T_c in the cuprate superconductors similar to the noncuprate. This may be a part of the understanding of the superconductivity of $(La_{1-x}Sr_x)_2CuO_4$ and $YBa_2Cu_3O_y$.

In conclusion, we observed the reproducible gap values in $(La_{1-x}Sr_x)_2CuO_4$ and $RBa_2Cu_3O_y$ (R=Y, Er, Gd). The observed tunneling densities of states are substantially the same as the BCS density of states. Observed double gaps at each material can be attributed to the gap anisotropies with respect to CuO_2 planes. The ratios $2\Delta/kT_c$ are 4.5–4.9 for $(La_{1-x}Sr_x)_2CuO_4$ and 3.3 and 4.2 for $RBa_2Cu_3O_y$ so that the significant difference of the ratio $2\Delta/kT_c$ between $(La_{1-x}Sr_x)_2CuO_4$ and $RBa_2Cu_3O_y$ was confirmed. These values remain constant although T_c decreases by changing x and y. These facts suggest no change of the pairing strength toward the antiferromagnetic insulating phases in both materials.

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