Observation of both pair and quasiparticle tunneling in intrinsic junction stacks fabricated on $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals

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We have investigated the tunneling properties of intrinsic Josephson junction stacks with various numbers N of CuO_2 bilayers fabricated on underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals. The effect of gap suppression based primarily on nonequilibrium superconductivity can be significantly reduced with a reduction in N. For an N as few as 40, both pair and quasiparticle tunneling characteristics are clearly observed. It is shown that a clear gap 2Δ of larger than 34 meV, the substantial conductance below the gap, and the I_cR_N product of less than 5 mV are the intrinsic features of tunneling along the c axis. Numerical calculation indicates that the quasiparticle current-voltage characteristics are consistent with the d-wave order parameter which is still suppressed by the nonequilibrium effect in a higher current region.

I. INTRODUCTION

The fabrication of superconductor-insulator-superconductor (SIS) tunnel junctions with high-T_c cuprates has been one of the greatest challenges, because they not only open up a route to electronic applications but also provide a direct probe into the order parameter and thus important information on the pairing mechanism in these materials. There have been enormous efforts to fabricate SIS, SIS′ (S′ is typically Pb), and SIN (N is normal metal) planar junctions mainly on the a-b plane of thin films or single crystals of, for example, $YBa_2Cu_3O_{7-\delta}$ (YBCO) (Refs. 1–3) and $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212).[4,5] However, no pair and quasiparticle tunneling characteristics which seem to reflect the intrinsic order parameter of these materials have ever been reported. This is primarily due to the very short coherence length of 0.1–0.2 nm along the c axis and the material's poor surface quality.

There have also been an increasing number of experiments on tunneling along the c axis of YBCO and Bi-2212 using a low-temperature scanning tunneling microscope (STM).[6-10] In many experiments, a substantial tunneling density of states (DOS) typically with a V shape below the gap 2Δ of 40–60 meV was observed and considered to be evidence for a highly anisotropic order parameter like that of a d-wave superconductor. However, the existence of the finite DOS near zero-bias voltage[6,7] as well as the anisotropic gap in the c axis or out-of-plane gap[1,8] is still controversial. The often observed anomalous background conductance structure and the problem of surface quality make it difficult to clarify these points and even evaluate the actual size of the order parameter. Furthermore, since the transverse momentum is not conserved upon tunneling because of the very small tip size and resultant uncertainty,[6] the tunneling conductance obtained using an STM does not strictly reflect the distribution of the order parameter in the k_x-k_y space. Thus the intrinsic properties of tunneling along the c axis of high-T_c cuprates are not sufficiently clear.

On the other hand, recent observation of intrinsic Josephson tunneling in small pieces of Bi-2212 single crystal[11,12] have strongly suggested that a stack of SIS Josephson junctions is naturally formed along the c axis in this material. Microwave emission experiments indicated that each CuO_2 bilayer stacked with a period of 1.5 nm acts as a superconducting electrode.[11,12] With these intrinsic junction stacks, we can expect to extract intrinsic information on the magnitude and the anisotropy of the order parameter, because of their planar structure and because their tunneling properties are much less sensitive to the structure and quality of the crystal surface than the case with measurements using a single planar junction or STM. However, clear quasiparticle tunneling properties have not been observed yet in Bi-2212 intrinsic junctions. Moreover, the 2Δ value of typically 20 meV previously estimated from the periodic voltage jumps in the current-voltage (I-V) characteristics[12] is much smaller than the values observed by STM measurements.[6,9,10] As we demonstrate below, the former problem mainly results from the fact that too many (typically 1000) junctions are included even in cleaved thin Bi-2212 single crystals.

In this paper, we report on the tunneling properties of intrinsic Josephson junction stacks with a controlled number of CuO_2 bilayers which are fabricated on underdoped Bi-2212 single crystals. For junctions with fewer than 100 CuO_2 bilayers, the effect of gap suppression which is primarily based on nonequilibrium superconductivity is substantially reduced and quasiparticle tunneling properties can be clearly observed. It is shown that these intrinsic tunneling properties are consistent with a d-wave symmetry of the order parameter.

II. EXPERIMENT

Bi-2212 single crystals were grown by a self-flux method using a platinum crucible.[13] Small pieces of these crystals with a typical size of $1\times1\times0.05$ mm^3 were cleaved and annealed in flowing oxygen at 600 °C for 60 h. The magnetically evaluated T_c of the annealed crystals was 79–83 K. After a 50 nm thick Au layer was evaporated on the surface of the crystals for electrical contacts, they were annealed again in O_2 at 600 °C for 1 h. Then these crystals were glued to Si substrates and junctions were fabricated using standard

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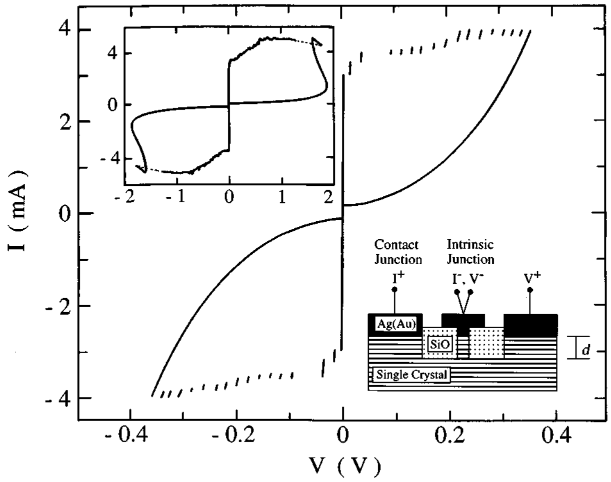

FIG. 1. I-V characteristics of a 300 nm thick junction stack at 11 K. The curve for a larger bias current is indicated in the inset. The critical current density j_c is 750–1250 A/cm^2. The schematic cross-sectional view of the junction stacks is also shown.

photolithographic and ion milling techniques. The junction thickness d and the number of included CuO_2 bilayers N were varied between 60 nm and 300 nm and between 40 and 200, respectively, with accuracy of $\pm10\%$ by adjusting the milling time. The self-aligned SiO insulating layer was deposited and finally an Ag wiring layer was formed.

The lateral size of the junctions was fixed at 20×20 μm^2 in the present experiments. The electrical contacts to the crystals were made via two contact junctions 200×200 μm^2 in size. Prior to the I-V measurements, the temperature dependence of the resistivity along the c-axis ρ_c was measured. A negative temperature dependence above T_c and a ρ_c value at room temperature of approximately 10 Ω cm clearly indicated that the crystals are in the underdoped regime. Since all the transport measurements were carried out using a three-probe configuration, a residual resistance was observed below T_c. However, this contact resistance was as small as 0.1–1 Ω and thus has no significant influence on the I-V measurements.

III. RESULTS AND DISCUSSION

Figure 1 shows the I-V curve at 11 K for a 300 nm thick junction stack. For a small bias current just above the critical current I_c, the junction exhibits a periodic voltage jump with a period V_j of approximately 20 mV and a relatively large hysteresis. These features and the switching between multiple quasiparticle branches are typically observed in a series array or stack of many SIS junctions with a substantial distribution of I_c. The V_j value was found to be independent of d and was 15–20 mV for other junctions. This value is quite similar to those previously reported by Kleiner *et al.*[12]

For a larger bias current, even larger hysteresis and negative resistance of the quasiparticle branch are observed. At a characteristic voltage V_c of approximately 1.8 V, each SIS junction contained in the stack is considered to be switched to the voltage state. This value is much smaller than the product of V_j and the number of SIS junctions in the stack $N=200$. This significant suppression of V_c has already been

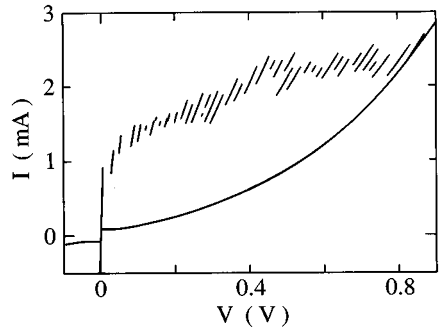

FIG. 2. I-V characteristics of a 60 nm thick junction stack at 4.2 K. The j_c is 250–500 A/cm^2. The V_c value is approximately 0.8 V.

reported in a junction stack of similar thickness.[12] As will be discussed later, neither V_j nor V_c/N is an actual gap of the CuO_2 bilayer. However, they are basically considered to be proportional to the gap.

Figure 2 shows the I-V curve for a 60 nm thick junction stack. Approximately 40 voltage jumps with a V_j of 20 mV can be observed. This explicitly indicates that SIS junctions are stacked with a period of 1.5 nm and thus each CuO_2 bilayer acts as an S electrode. The V_c value is approximately 0.8 V. Thus V_c/N is very close to V_j in this stack.

In Fig. 3, the normalized V_c value is plotted as a function of N for junction stacks with a similar j_c of approximately 500–1000 A/cm^2. The suppression of V_c is more significant as N is increased. As was previously pointed out, this suppression might be due to joule heating. However, suppression of greater than 55% in the present experiments and 85% reported for thicker junction stacks[12] cannot be explained only by the heating effect.[14]

Another possible explanation for the suppression of V_c is nonequilibrium superconductivity due to quasiparticle injection.[15,16] Although our knowledge of the pairing mechanism and physical properties such as quasiparticle recombination time is still lacking, we could roughly estimate its

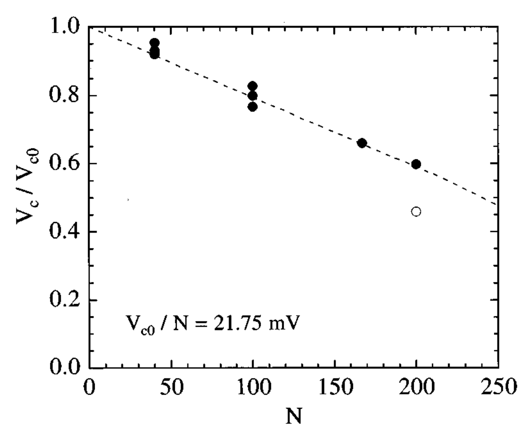

FIG. 3. Variation in characteristic voltage V_c with number of CuO_2 bilayers, N, contained in different junction stacks with a j_c of approximately 500 A/cm^2 (filled circles) and 1000 A/cm^2 (open circle). The V_c value is normalized by $V_{c0} = 21.75 \times N$ mV. The broken line is the variation roughly estimated assuming nonequilibrium gap suppression based on the T^* model and a current density J of 1000 A/cm^2 (see text).

effect on the gap suppression as follows. Assuming that each CuO_2 bilayer with the thickness t_s of 0.3 nm is homogeneously injected by quasiparticles with the density $I_{qp} = J/et_s$, the normalized excess quasiparticle density n in each CuO_2 bilayer is, to the first approximation, given by, $n = I_{qp}\tau_{eff}/4N(0)\Delta$.[15] Here, $N(0)$ is the one-spin density of states and τ_{eff} is the effective quasiparticle recombination time which is expressed as $\tau_{eff} = \tau_R(1+\tau_{es}/\tau_B)$. τ_R and τ_B are the intrinsic quasiparticle recombination time and phonon pair-breaking time, respectively. τ_{es} is the phonon escape time which is given by $\tau_{es} = 4d/\eta v_s$. Here, the thickness d can be assumed to be identical to the thickness of the junction stack. η and v_s are the transmission probability of phonons at the junction-crystal boundary and the sound velocity in the stack, respectively.

Since τ_{es} is usually much larger than τ_B in particular for junction stacks, $\tau_{eff} \approx \tau_{es}\tau_R/\tau_B$ holds and thus τ_{eff} and n can be considered to be approximately proportional to d or N. In Fig. 3, the dependence of gap suppression on N given by the T^* model[16,17] is compared with the experimental data using τ_{eff} as a variable parameter. The fitted curve gives a τ_{eff} of 4 ns for a 300 nm thick stack. This value is only a factor of 5 larger than the τ_{es} of 0.8 ns, which is estimated assuming a v_s of 5×10^5 cm/s,[18] η of approximately 0.3 and $4N(0)\Delta$ of 4×10^{21} cm^{-3}, and may be within an acceptable range.

Figure 4 (a) shows the I-V curve extended to a higher voltage range for the same junction stack as in Fig. 2. A few notable features can be recognized here. First of all, a clear gap structure is observed at a voltage V_g of 1.35 V which is much larger than V_c. The I-V curves at various temperatures are shown in Fig. 4(b). All the curves coalesce into one linear line at higher voltages and the gap structure disappears above T_c. This excludes other possibilities such as a resistive transition in the contact junctions. Thus we can define $2\Delta_{obs} = V_g/N$ as the gap size which is approximately 34 meV. We notice that V_g shows a substantial temperature dependence even at $T < 10$ K, as seen in the inset. This indicates that the gap is still suppressed to some extent by the injection of a quasiparticle current with a density of approximately 4 kA/cm^2. Thus the value of 34 meV provides only the lower gap size limit. It is also noted that the normal tunneling resistance for each junction R_N of approximately 2.2 Ω is nearly temperature independent.

The second noticeable feature is the substantial conductance below V_g. This excess conductance depends strongly on the bias voltage and therefore cannot simply be attributed to the increase in the nonequilibrium quasiparticle density. Another feature of particular interest in Fig. 4(a) is that the Josephson current seems to be significantly suppressed. The I_cR_N product for each junction is 4–5 mV which is less than 1/5 of the BCS value $\pi\Delta_{obs}/2e$. In a conventional planar Josephson junction, a substantial suppression of I_c is also expected if the junction size is larger than the Josephson penetration depth, λ_j.[19] However, in a Josephson junction stack with an S layer much thinner than the magnetic penetration depth λ, the characteristic length which defines the long junction limit is given by the screening length, $\lambda_m = [\Phi_0/2\pi\mu_0(t_s+t_i)j_c]^{1/2}$, rather than λ_j.[20] Here, t_i is the thickness of the spacing between adjacent CuO_2 bilayers, which is 1.2 nm for Bi-2212. A j_c value of 1 kA/cm^2 thus

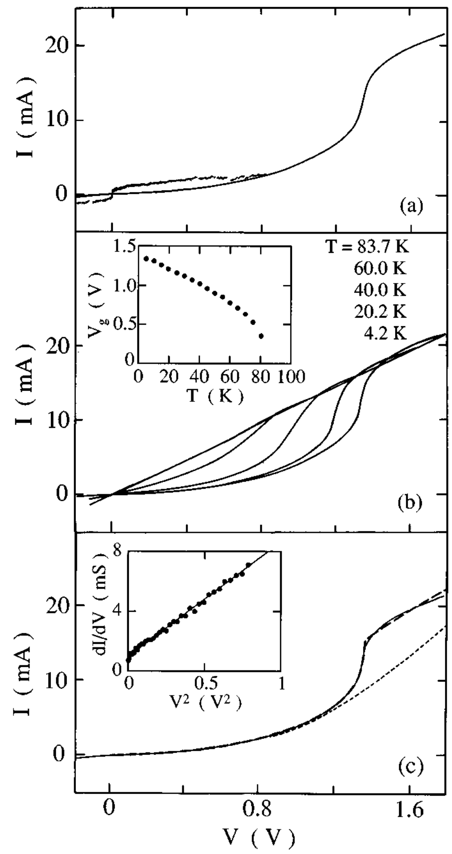

FIG. 4. I-V characteristics extended to a higher voltage for the same junction stack as in Fig. 2. (a) I-V characteristics at 4.2 K. (b) Temperature dependence of quasiparticle tunneling characteristics. The normal tunneling resistance for each junction, R_N, is approximately 2.2 Ω. The temperature dependence of gap voltage V_g is shown in the inset. (c) Comparison of the quasiparticle tunneling curve at 4.2 K (thin solid line) with theoretical curves assuming a $d_{x^2-y^2}$ symmetry of the order parameter in the k_x-k_y space. The broken and dotted lines are curves calculated with and without taking account of the nonequilibrium effect (see text), respectively. The existence of a small parallel conductance (6.5% of $1/R_N$) is also assumed. $2\Delta_0 = 50$ meV and $p = 1.0$ give the best fit. The inset shows the observed conductance for smaller voltages which exhibits a clear V^2 dependence.

gives a λ_m value of 150 μm, suggesting that the present junction stacks act as short junctions. It should also be noticed that both the reduced Josephson current and the substantial conductance below the gap give rise to the large difference between $2\Delta_{obs}$ and the V_j value of 14–20 mV.[21] Thus, the similarity of the V_j value observed in the present junction stacks as well as the stacks studied by Kleiner *et al.*[12] supports that the reduced I_cR_N product and the substantial conductance below the gap voltage are the *intrinsic features* of tunneling in intrinsic junction stacks based on Bi-2212.

In order to make the large subgap conductance much clearer, the derivative conductance dI/dV is plotted against V^2 in the inset of Fig. 4(c), indicating that dI/dV is proportional to V^2. This particular dependence implies that the quasiparticle density of states $N(E)$ is linear in the excitation energy E near the Fermi level. Here, it is important to notice that such $N(E)$ can be expected in a two-dimensional (2D) d-wave superconductor.[22] Therefore, for comparison, we numerically calculate I-V curves based on the 2D pure d-wave order parameter

$$\Delta(\theta)=\Delta_0\cos(2\theta), \tag{1}$$

where $\theta=\tan^{-1}(k_y/k_x)$. The normalized quasiparticle density of states $N(E)$ for tunneling along the c axis is then given by

$$N(E)=\mathrm{Re}\int_0^{2\pi}\frac{1}{2\pi}\left[\frac{E}{\sqrt{E^2-\Delta^2(\theta)}}\right]d\theta. \tag{2}$$

The tunneling quasiparticle current I_{qp} is calculated by

$$I_{\mathrm{qp}}(V)=\frac{1}{R_N}\int_{-\infty}^{\infty}N(E)N(E-eV)\{f(E-eV)-f(E)\}dE, \tag{3}$$

where $f(E)$ is the Fermi function. Since the inset of Fig. 4(c) indicates the existence of a small parallel conductance (approximately 6.5% of $1/R_N$) probably of extrinsic origin, I-V characteristics are expressed as

$$I=(0.065/R_N)V+I_{qp}(V). \tag{4}$$

The dotted line in Fig. 4(c) is the fit to the observed curve at 4.2 K. Although the fit is pretty good for $V<1.2$ V, a significant deviation is seen near the gap voltage. As mentioned above, this is likely due to the effect of the nonequilibrium superconductivity by self-injection of quasiparticle current. In order to take this effect into account, we further replace Δ_0 in Eq. (1) by

$$\Delta(J_{\mathrm{qp}})=\Delta_0(1-pJ_{\mathrm{qp}}^n) \quad (n=1,2), \tag{5}$$

which is similar to the T^* model employed to explain the data of Fig. 3. Here, $J_{\mathrm{qp}}=I_{\mathrm{qp}}/I_0$ is the quasiparticle current normalized by $I_0=2\Delta_0/eR_N$ and the variable parameter p represents the degree of nonequilibrium effect. The exponent n determines the way how quasiparticle current suppresses the gap. Since, according to the T^* model, the square rather than linear dependence of Δ on J_{qp} may be more appropriate for a significant quasiparticle injection, $n=2$ is employed.[23] The resulted fit is shown by the broken line in the figure. The fit is excellent over the whole V range measured in particular $V<V_g$, indicating that the quasiparticle I-V characteristics of the present intrinsic junction stack are consistent with the d-wave order parameter.[24] It is noted that the deduced $2\Delta_0$ value of 50 meV agrees with those observed by STM measurements.

IV. CONCLUSION

We have shown that the effect of nonequilibrium superconductivity on the I-V characteristics of intrinsic Josephson junction stacks based on Bi-2212 can be significantly reduced as the number of the CuO_2 bilayers N is decreased. For N as few as 40, both pair and quasiparticle tunneling characteristics become clearly visible. It is shown that a clear gap of larger than 34 meV, substantial excess conductance below the gap, and a substantially reduced I_cR_N product are the intrinsic features of tunneling along the c axis between the CuO_2 bilayers. The observed quasiparticle tunneling curve is well explained by assuming an order parameter with a d-wave symmetry in the k_x-k_y space and taking account of the nonequilibrium effect which still exists for higher bias currents.

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