Dynamic Cooper-pair breaking by tunnel injection of quasiparticles into a high-T_c $YBa_2Cu_3O_7$ superconductor

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The measurements on strong quasiparticle injection through a tunnel barrier into a high-T_c $YBa_2Cu_3O_7$ (YBCO) bicrystal film/junction or a YBCO film are presented. With an increase of tunnel injection current, both the Josephson and film critical currents decreased and were driven to zero to yield the normal state at a certain critical injection current density which ranged 20–140 A/cm^2 for 70 to a few 100 mV voltage bias. The results are qualitatively different from simple heating phenomenon and reflect nonthermal nature of high-T_c superconductors under injection.

The quasiparticle injection phenomenon into a nonequilibrium superconductor received considerable attention for low-T_c superconducting materials.[1–4] The quasiparticle injection is held either by optical excitation or tunnel injection of quasiparticles or phonon injection. Among them, the intense tunnel injection yielded a variety of interesting phenomenon such as the gap suppression,[5–8] the instability toward the spatial inhomogeneous states[9] and the multiple gap structures,[10–12] and the possible applications to electronics.[13,14] The quasiparticle injection into a high-T_c superconductor has been so far held by optical excitation technique.[15–19] It has been reported that both bolometric and nonbolometric (nonequilibrium) responses were obtained for a high-T_c superconductor, however, their essential natures are still controversial. The observed quasiparticle recombination time was 2–5 psec.[15] In the case of a high-T_c superconductor, because of its large gap parameter and high T_c, it is difficult to drive it to a strongly perturbed nonequilibrium state. In fact, there are very few reports on this matter.

In this paper, we report the measurements of tunnel injection of quasiparticles into a $YBa_2Cu_3O_7$ (YBCO) film and a YBCO bicrystal film/junction.[20] In contrast to optical excitation (excitation energy 1 eV), tunnel injection corresponds to injection of quasiparticles with energy 20 meV to a few 100 meV. The quasiparticle injection through a tunnel barrier led to clear reduction of bicrystal Josephson current and YBCO film critical current. At a certain injection current, the supercurrent completely disappeared to yield the normal state at 4.2 K. The critical injection current density ranged 20–140 A/cm^2. It was found that the observed phenomena were qualitatively different from simple heating phenomenon and were considered to be of essentially nonequilibrium nature.

We have used two types of sample geometries as shown in Fig. 1. One is an Au/MgO/YBCO junction grown on a $SrTiO_3$ (100) bicrystal substrate (Dowa Mining Co. Ltd.) (a) and the other is that grown on a MgO(100) substrate. The crystal angle at the interface of the bicrystal substrate was 24°. Sample (a) contains a bicrystal Josephson junction. The samples were fabricated by *in situ* sequential deposition of trilayer films utilizing an electron-beam and resistive heater coevaporation chamber equipped with a metal mask changing system. In both samples, YBCO films were *c*-axis oriented. The thicknesses of YBCO, MgO, and Au films were 70, 4, and 70 nm, respectively. The junction had a cross-type geometry and the injection area was 0.2×0.2 mm^2. The junction resistance was typically 5–10Ω. We have tested eight samples all of which exhibited qualitatively similar behavior and here present the representative data.

We first present the measurements on an Au/MgO/YBCO trilayer on a $SrTiO_3$ bicrystal substrate in Fig. 1(a). Figure 2 shows the observed *I-V* characteristics of a Josephson bicrystal junction under microwave irradiation of 11 GHz. The appearance of Shapiro steps and large reduction of supercurrent with an increase of microwave power intensity assure that the supercurrent was dominantly of Josephson nature. The magnetic field produced by transport current along an Au film also caused the modulation of supercurrent. We believe that Josephson junction will be formed at the bicrystal boundary since the current-voltage characteristics of YBCO films deposited on single-crystal MgO or $SrTiO_3$ substrates by the same technique never yielded Shapiro steps under microwave irradiation even if the film critical current was small. For the Au/MgO/YBCO injector tunnel junction, its I_{inj}-V_{inj} characteristic is shown in the inset of Fig. 3. The gap structure was smeared as usually observed for a YBCO tunnel junction[21–23] but the current rising behavior at the gap edge (at about 20 mV)

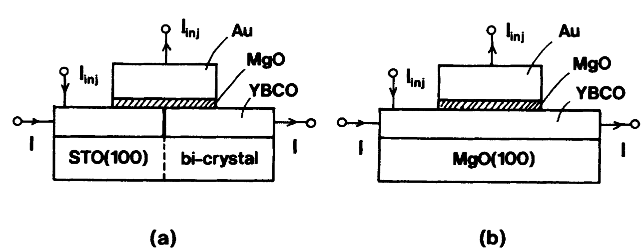

FIG. 1. Sample geometries for tunnel injection of quasiparticles into a YBCO bicrystal film/junction (a) and a YBCO film (b) where I_{inj} is the injection current and I is the transport current along a YBCO film.

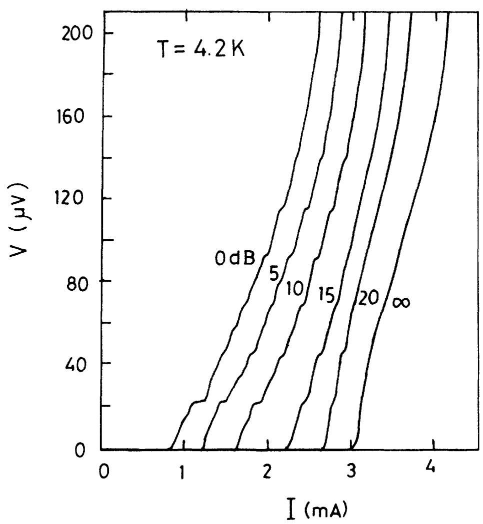

FIG. 2. Current-voltage characteristics of a bicrystal junction under microwave irradiation of 11 GHz.

was clear. The smeared gap structure may be attributed to the presence of excess current due to the distributed normal regions caused by oxygen deficiency on the surface of a YBCO film.

Figure 3 shows the Josephson I-V characteristics of a YBCO film under the influence of injection current. In this measurement, two currents were fed in a YBCO film; one is the injection current $I_{\rm inj}$ and the other is the transport current I through a YBCO film as shown in Fig. 1. The injection current was fed from the Au film to the YBCO film. First, we mention about the case that $I_{\rm inj}$ and I flew in the same direction in a YBCO film, corresponding to the positive current axis in Fig. 3. The injection current flew from the Au film to the YBCO film. It is noted here that the quasiparticle-injection effect did not depend on the polarity of the injection current. For $I_{\rm inj}$ less than 12 mA, the Josephson critical current $I_{\rm jc}$ did not change greatly. For $I_{\rm inj}$ greater than 12 mA, $I_{\rm jc}$ decreased rapidly with an increase of injection current and became zero at 14 mA. We define this critical injection current as $I_{\rm ic}$. The injection voltage at $I_{\rm inj}=I_{\rm ic}=14$ mA was 94 mV, corresponding to the maximum injected quasiparticle energy of about 5Δ. The dissipated power was about 3 W/cm^2 at 14 mA, which is only a few times greater than that of low-T_c Pb superconductors.[5] For further increase of $I_{\rm inj}$, the voltage developed even at $I=0$. The fact indicates that the injected area became actually normal due to injection of quasiparticles.

Figure 4 shows the dependence of Josephson critical current $I_{\rm jc}$ on the injection current $I_{\rm inj}$ when $I_{\rm inj}$ and I flew in the same direction in the YBCO film. In the case that $I_{\rm inj}$ was less than 12 mA, small modulation of $I_{\rm jc}$ was observed. The modulation behavior was mostly attributed to the effect of magnetic field produced by injection current $I_{\rm inj}$ itself since a current passing through a Au film only also yielded a similar modulation effect on $I_{\rm jc}$. Hence, for $I_{\rm inj}<12$ mA, the result is consistent with the combined effects of self-induced magnetic field and quasiparticle injection. The strong suppression of $I_{\rm jc}$ for $I_{\rm inj}>12$ mA was due to heavy injection of quasiparticles. It is noted that the strong suppression of $I_{\rm jc}$ was not observed simply by feeding a current along the Au film only even up to 40 mA. The current ratio $I_{\rm jc}/I_{\rm ic}$ was about 0.3 and local current gain $dI_{\rm jc}/dI_{\rm inj}$ near $I_{\rm ic}$ was about 5. The observed behavior was qualitatively different from that expected for the simple heating model and is considered to be of nonequilibrium nature. The injected quasiparticles through a tunnel barrier into a high-T_c YBCO superconductor relax toward the gap edge by emission of phonons and recombine to form Cooper pairs at gap edge by emission of 2Δ phonons. The resultant phonons with energy greater than 2Δ again contribute to pair breaking to excite quasiparticles and these processes continue until some steady nonequilibrium state is established.

Next, when $I_{\rm inj}$ and I were in the opposite direction (corresponding to negative current axis in Fig. 3), the

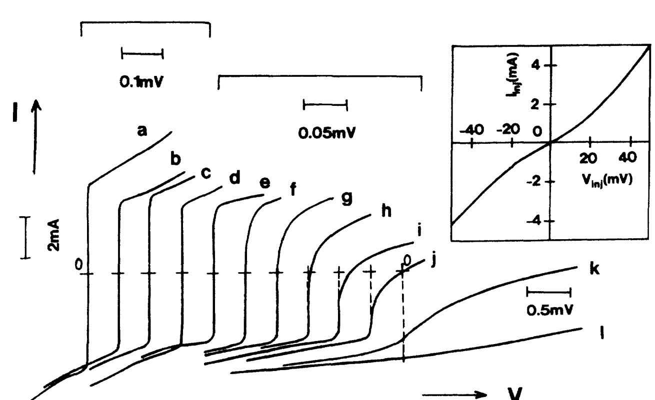

FIG. 3. Current-voltage characteristics of a bicrystal junction at $T=4.2$ K at different injection currents $I_{\rm inj}$ where $a-1$ corresponds to $I_{\rm inj}=0$, 6.0, 10.0, 12.0, 13.0, 13.6, 14.0, 14.4, 15.0, 15.4, 18.0, and 20.0 mA, respectively. Note that the voltage axis of each characteristic was displaced slightly. The inset shows the current-voltage characteristic of the Au/MgO/YBCO injector tunnel junction.

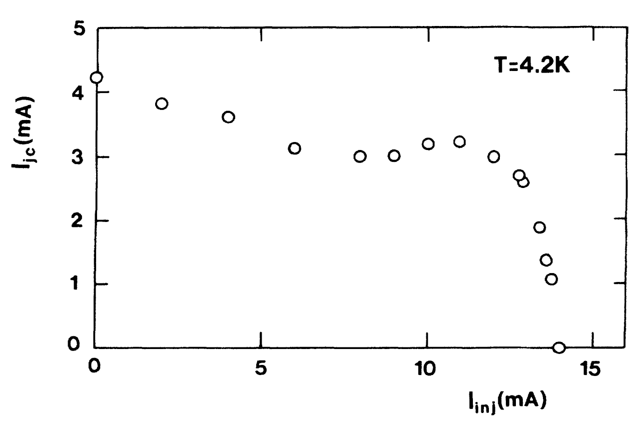

FIG. 4. Josephson critical current of a bicrystal junction as a function of injection current $I_{\rm inj}$.

behavior for I_{inj} below 12 mA was similar to the case stated above. For stronger injection (I_{inj} greater than 14 mA), the great change in the *I-V* curves was observed. The injection effect was the strongest when $I=0$ in this case and for a finite value of I, it was weakened and recovered zero voltage state for a certain range of current I because of opposite current direction. But when I_{inj} is sufficiently large and becomes above a certain threshold, only the normal state resulted in, yielding near-ohmic *I-V* characteristics.

As described above, the asymmetric *I-V* characteristics appeared because two currents, I_{inj} and I, were involved. The generation of voltage at $I=0$ was due to injection current I_{inj} itself, which was given by $R_{eff}I_{inj}$, where R_{eff} is the effective film resistance caused by quasiparticle injection. By assuming the YBCO film normal resistance in the junction part as R_n, the value of R_{eff} increased from zero to the final normal value close to $R_n/2$ by the analysis of the four-terminal method.[5]. For example, for $I_{inj}=20$ mA, R_{eff} became about 5Ω, which was large due to high resistivity of YBCO material. The voltage at zero transport current will be reduced considerably provided that the width of the Au film becomes much smaller since the injection phenomenon depends on the current density.

The observed critical injection current I_{ic} (14 mA) was much greater than I_{jc} at $I_{inj}=0$ (4.2 mA). Hence, we consider that the flow of injection current in the junction region was quite asymmetric, allowing the supercurrent across the bicrystal junction below I_{jc} and the remainder current flew through half side of the junction area without passing through the bicrystal junction. In fact, this was confirmed by the measurement with feeding I_{inj} only and observing the voltage onset in a YBCO film. The voltage appeared at $I_{inj}=I_{ic}$ much greater than the value of I_{jc}. The behavior in Fig. 4 may be also interpreted by an asymmetric flow of I_{inj}. With increase in I_{inj}, the current extraction side of the bicrystal junction is strongly perturbed and the gap is largely reduced, while the other side is little perturbed. Suppose that the nonequilibrium state is described by some effective temperature T^*, then the dependence of I_{jc} on T^* corresponds to the case of large gap difference between two electrodes, which exhibits strongly nonlinear behavior against T^* in the framework of Ambegaokar-Baratoff's theory.[24] Since $T^*$is roughly proportional to $I_{inj}^{1/2}$ in the high-voltage injection regime, the behavior of I_{jc} against I_{inj} will also exhibit similar nonlinear behavior consistent with the result of Fig. 4. The origin of an asymmetric flow probably arises from minimization of free energy in the perturbed YBCO film. As far as the film is in the superconducting state, its free energy will be smaller than that of the dissipated state accompanying voltage generation.

To investigate the nonequilibrium effect by tunnel injection of quasiparticles directly, the measurement using the geometry in Fig. 1(b) without containing a bicrystal junction was also performed. The T_c of a YBCO film was 75 K. Figure 5(a) depicts the measurement on the film critical current I_c vs the tunnel injection current I_{inj} when the direction of current flow for I_c and I_{inj} were the same at 4.2 K. Without quasiparticle injection, I_c was 10 mA (0.8×10^5 A/cm^2). In this case, I_c was not perturbed by the transport current through a Au film. I_c decreased monotonically with an increase of I_{inj} and became completely zero at $I_{inj}=I_{ic}=8$ mA. The dissipated power at 8 mA corresponding to the junction current density 20 A/cm^2 was only 1.4 W/cm^2, the same order of magnitude as the case of low-T_c superconductors.[14] We note that the other sample with better film critical current $I_c=70$ mA (5×10^5 A/cm^2), I_{ic} was 55 mA corresponding to the junction current density of 140 A/cm^2. Figure 5(a) shows that the current gain I_c/I_{ic} was greater than 1. Below 1.5 mA, the injection voltage V_{inj} was less than Δ/e, hence quasiparticle injection was not held and I_c remained almost unchanged. The enhanced reduction of I_c above $I_{inj}=6$ mA ($eV_{inj}\approx3\Delta$) may be attributed to the extra pair-breaking effect by relaxation phonons with energy greater than 2Δ. We note that the observed *I-V* characteristics were also qualitatively similar to Fig. 3, in which the asymmetric behavior against the direction of transport current I for a fixed direction of I_{inj} was again seen. This together with the result in Fig. 3 assures that the observed phenomenon may originate from the nonequilibrium injection effect in a YBCO film itself. The dashed line in Fig. 5(a) corresponds to the theoretical curve as expected from the simple heating model, in which tunnel injection merely raises the film temperature. The steady-state temperature is determined by heat input and thermal boundary resistance. For simplicity, we have assumed here that bath temperature T is zero because of very low reduced temperature ($T/T_c=0.058$) and the film critical current is given by the expression

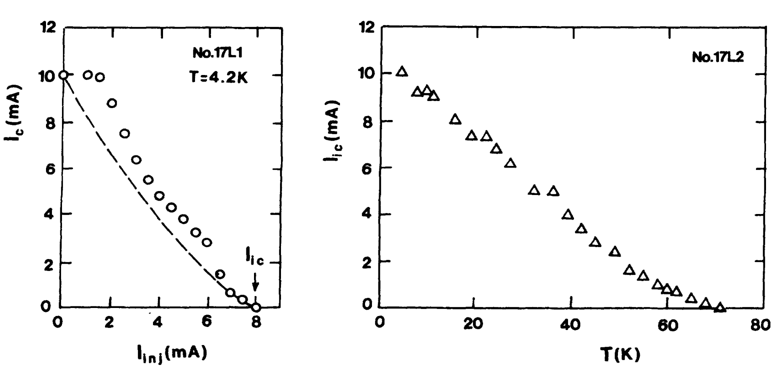

FIG. 5. (a) YBCO film critical current I_c as a function of injection current I_{inj}. The dashed line corresponds to the calculated curve for a simple heating model. (b) Temperature dependence of critical injection current I_{ic}.

$I_c = I_{c0}(1-t^{*2})^{3/2}(1+t^{*2})^{1/2}$ for the effective temperature T^* ($t^* = T^*/T_c$), where I_{c0} is the unperturbed critical current at $T=0$, an approximate formula valid for all temperatures.[25] It is clear that the observed data appreciably deviated from the simple heating curve except near I_{ic}. The behavior in Fig. 5(a) was different from that in Fig. 4, which arises from the different supercurrent nature of Josephson critical current I_{jc} and film critical current I_c. Figure 5(b) shows the temperature dependence of critical injection current I_{ic}, which is nearly linear against temperature. It is pointed out that threshold I_{ic} was clearly defined even close to T_c. We also note that the measurements using Pb/MgO/YBCO junctions with stronger nonlinear tunnel characteristics including almost ideal Pb gap structures yielded qualitatively similar behavior to those of Au/MgO/YBCO junctions, whose large body of work will be published separately.

We consider that the above-observed phenomena arise from both the quasiparticle injection effect and the pair-breaking effect by the transport current in a film since the magnitudes of I and I_{inj} were rather close, although the observed I-V curves were quite different from those expected from the current pair-breaking effect only. In case that I and I_{inj} flow in the same direction in the film, the measured critical current I_c^{meas} is given by

$$I_c^{meas} = I_c(I_{inj}) - I_{inj} = I_c(0) - I_{inj} - \Delta I_c$$

where $\Delta I_c = I_c(0) - I_c(I_{inj})$. The term ΔI_c is solely due to the quasiparticle injection effect. By assuming the expression of film current density $J_c = H_0/4\pi\lambda(0)$ at low reduced temperatures where H_0 is the thermodynamic critical field and $\lambda(0)$ is the London penetration depth at $T=0$, the decrease in I_c [$a = (I_c - \Delta I_c)/I_c < 1$] corresponds to the gap reduction of $\delta\Delta = (1-a^{2/3})\Delta_0$ using the relation $H_0^2/8\pi = N(0)\Delta_0^2/2$. Accordingly, the excess quasiparticle number density n in units of $4N(0)\Delta_0$ is approximately given by $n \sim (1-a^{2/3})/2$ using the relation $\Delta/\Delta_0 = 1-2n$ given by the μ^* and T^* models[26,27] since the observed value of a is 0.5–0.8 depending on the samples.

The calculated value of J_c is 5×10^8 A/cm^2 by assuming $H_0 = 1$ T and $\lambda(0) = 150$ nm, the value applies to the ideal thin films for which strong superconductivity is maintained all over the sample volume. In case of high-T_c superconductors, however, I_c strongly depends on the sample conditions according as the oxygen deficiency, etc. In fact, J_c of our samples was $(0.5-5)\times10^5$ A/cm^2 far below the theoretical value. The reduction of current density means that the effective Cooper-pair number density, i.e., the number of Cooper pairs simply divided by the sample volume, is small in actual samples. This effect can be taken into account, equivalently, by considering that the sample volume $w_1 w_2 d$ is effectively reduced by an amount $b = I_c^{meas}/I_{c0}^{calc}$ which should be included in the following calculation, where w_1 and w_2 are the widths of YBCO and Au films, respectively, and d is the YBCO film thickness.

Now in the nonequilibrium steady state, the total quasiparticle number density N_{eff} is given by $N_{eff} = I_0\tau_{eff}$ using the Rothwarf-Taylor equation,[28] where I_0 is the injection rate and τ_{eff} is the quasiparticle relaxation time. If we assume the relation $I_0 = FR_n I_{inj}^2/2\Delta_0 b w_1 w_2 d$ between I_0 and I_{inj} given by Wong *et al.*[13] where R_n is the junction normal resistance, F is the conversion factor and $N_{eff} \sim N(0)\Delta_0$, then

$$\tau_{eff} \sim 4(1-a^{2/3}) b N(0)\Delta_0^2 w_1 w_2 d / FR_n I_{inj}^2 .$$

Substituting a set of parameters $R_n I_{inj}^2 \sim R_n I_{ic}^2 \sim 1-10$ mW, $N(0)\Delta_0 \sim 10^{21}$ states cm^{-3}, $\Delta_0 \sim 10-20$ meV, $F \sim 1$, $w_1 = w_2 = 0.02$ cm, $a \sim 0.5-0.8$, and $b = (0.1-1)\times10^{-3}$ yields $\tau_{eff} \sim 0.1-1$ nsec whose order of magnitude is consistent with the recent measurements by optical excitation.[18] The consideration of quasiparticle injection into two-dimensional CuO_2 planes only yields another reduction of τ_{eff} by a factor of about $\frac{1}{3}$. The quasiparticle injection effect becomes much more dominant as compared with the current pair-breaking effect if the width w_2 of Au is made much smaller, contributing to large current gain, which we leave for further investigation.

In summary, we have reported the measurements on tunnel injection of quasiparticles into high-T_c superconducting YBCO films. The Josephson critical current and the film critical current decreased with an increase of tunnel injection current and became zero to yield the normal state at a certain critical current I_{ic}. The current gain was greater than 1 for the latter. The observed phenomenon was clearly different from that expected from simple heating model and essentially of nonequilibrium nature, which will serve as investigations of the fundamental properties of both superconducting and normal states in high-T_c superconductors. Incidentally, both the large current gain and the small dissipated power may be attained by making the junction size smaller, so that it is possible to develop a quasiparticle injection-controlled three-terminal device of μw dissipation.

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