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Photo-induced Resistive State and Effective Quasiparticle Lifetime in Nonequilibrium Superconducting Sn Films Near Transition Temperature

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Abstract

The photoinduced resistive state in superconducting Sn films was measured by means of conventional dc-method, and the effective quasiparticle lifetime was deduced from the experimentally observed decay time of quasiparticles. Near transition temperature, T_c, the mean lifetime of phonon for creating quasiparticles, τ_{pq}, is dominant in quasiparticle relaxation. It shows that τ_{pq} increases with the decreasing temperature. The current-voltage characteristics are shown to be nonlinear under illumination. These are not explained in terms of simple lattice heating or the modified heating theory and suggest that a spatially inhomogeneous state would be established in the optically excited superconducting Sn films.

1. Introduction

Recent experimental[1~4)] and theoretical[5~7)] studies on the superconducting films driven far from equilibrium state have brought about many interesting results. The most interesting feature is a wider phase-transition from superconducting to normal state[3,4)]. This effect would not be only due to a simple lattice heating, but to an excess number of quasiparticles created by the external field. Various theoretical suggestions for this phenomenon have been proposed. The quasiparticle distribution affects the superconducting energy gap through an ambient temperature and the chemical potential. Owen and Scalapino[5)] have investigated a system in which the quasiparticles attained thermal equilibrium with respect to the lattice, but were not in chemical equilibrium with Cooper pairs (μ^* model). They have shown that the μ^* model for a nonequilibrium superconductor led to a first-order phase transition to the normal state at a certain excess quasiparticle density and exhibited an inhomogeneous state at low temperatures consisting of separate superconducting and normal regions, or regions with different gaps. Parker[6)] has treated the system in terms of high energy phonons greater than twice the gap (T^* model) and then the superconductor undergoes a second-order phase transition as the quasiparticle density increases (These are shown in Fig. 1).

Some time ago, Rothwarf and Taylor[8)] pointed out the importance of excess phonons on the quasiparticle recombination lifetime in a nonequilibrium superconductor. They derived an equation which represented the relation between the apparent qua-

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siparticle recombination time obtained experimentally and the intrinsic recombination time. In order to determine the intrinsic recombination time, some experiments[9,10] have been carried out in a nonequilibrium superconductor.

In a region of low temperatures compared with T_c, Levine and Hsieh, etc.[9,10] showed that the recombination time is roughly proportional to exp (Δ/k_BT), where k_B is Boltzmann constant, Δ the energy gap and T the absolute temperature. This is caused by the fact that the number of excited quasiparticles available for paring with a given quasiparticle is propotional to exp $(-\Delta/k_BT)$. However, upon approaching T_c $(\Delta/k_BT\sim 2)$, the recombination time deviates from the above dependence and a clear account for this phenomenon has not been given.

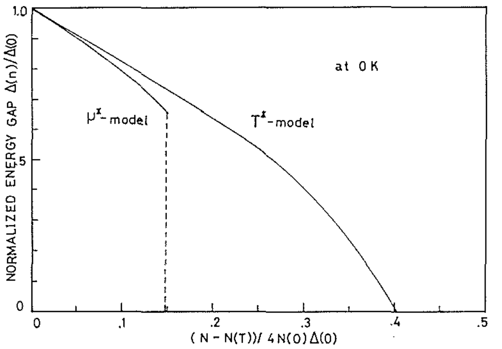

Fig. 1. Normalized energy gaps for the Owen-Scalapino μ^* model and Parker T^* model as a function of normalized excess quasiparticle density, where $n=(N-N(T))/4N(0)\Delta(0)$. For Sn, $4N(0)\Delta(0)=3.29\times10^{19}\,cm^{-3}$.

The purpose of the present work is to investigate a dc-resistive state in superconducting Sn films induced by CO_2 laser light and to measure the time dependence of quasiparticles near T_c. The principle of this experiment can be described as follows. Quasiparticles are produced by pair breaking with illumination of laser light. After being excited, a quasiparticle may (1) emit phonons or (2) give to other quasiparticles its exciting energy and relax to an energy level near the Fermi surface. Then it may recombine with another quasiparticle with emission of a phonon. While the free energy in the superconductor is larger than or equal to that of normal conductor temporally or spacially, this will give rise to a finite resistance. The measured resistance and its decay time provide information of quasiparticle lifetimes and the relaxation process.

2. Experimental Procedure

Films were obtained by evaporating of high purity Sn (99.999%) at the rate of 1∼5 Å/sec onto polished sapphire or cover glass substrates at room temperature. Thicknesses of films were 400 to 2000 Å. The electric resistivity at 300 K $(\sim 3\times 10^{-5}$ ohm·cm) was close to the bulk value. Resistance ratios R(300 K)/R(4.2 K) were 6 to 8.

Sample shapes suitable for this method were obtained by scribing away thin lines of Sn films on the substrate surface. Sample dimensions were usually $0.2\times 24\,mm^2$. Parts of electrodes contacted by indium solder were masked for laser light. The reverse side of the substrate was attached to a copper block with Apiezon-N grease. Sample and Ge-thermometer inserted into the copper block were set in a vacuum can. The detail of arrangement is shown in Fig. 2.

A CO_2 laser used in this experiment was multimode at 10.6 μm, and 22 W in maximum. The outputs were continuous wave with spot of 6 mm in diameter. Laser light was chopped by a mirror rotating at 450 Hz. An optical loss of 25%, for chopper, mirror, and Ge prism with antireflecting coating is estimated.

For tin, the Fermi velocity v_f is 0.65×10^8 cm/sec and the London penetration depth λ_L is 355 Å. Optical penetration depth for CO_2 laser ($\omega \sim 1.78\times 10^{14}$ rad/sec) is approximately equal to 210 Å calculated from an anomolous skin effect $\sim 1.26\times \omega^{-1/3} v_f^{1/3} \lambda_L^{2/3}$. Therefore, in this experiment the optical penetration depth was less than the film thickness.

The resistive response for the laser pulse was measured by a constant current method using coaxial cables. The film resistance induced by the laser pulse was determined from the difference between a peak of the response pulse and the background. The T_c in zero light was determined from the temperature in which the electrical resistance of the film became a half value of normal resistance.

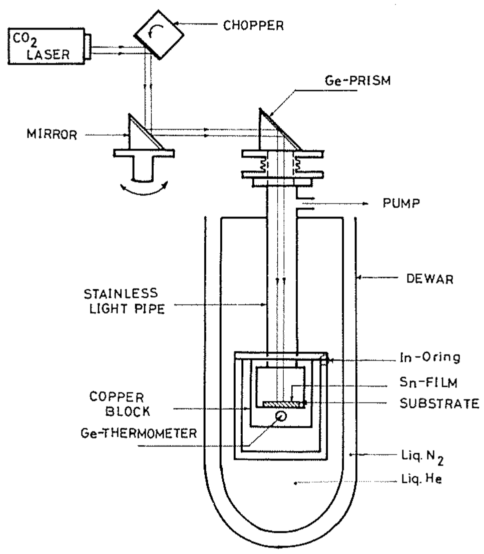

Fig. 2. Experimental arrangement.

3. Photo-induced Resistive State

The value of T_c in zero light was 3.87 K on the average with a variation of less than 0.03 K from sample to sample. T_c had a tendency to decrease with increasing current from 400 to 800 μA, but the magnitude of change in T_c was very small, and remained constant within a few millidegrees. Although a width of the superconducting transition in zero light is only a few tenmillidegrees, the photo-induced resistive state shows very broad phase transition to normal state. This is shown in Fig. 3. Here resistive ratio to normal state resistance, R/Rn, is shown as a function of the initial temperature, where R is sum of the resistance in zero light and the resistance induced by illumination.

Lattice heating by laser light causes a depression of T_c. In view of thermal effect, nonuniformity of the light absorption over the film thickness will be small, since the thermal diffusion time in Sn films (10^{-8} sec) is short compared with pulse width (typically 5μsec). Therefore, the thermal effect will not have influence on the configuration of S-N transition curve in zero light. We have estimated the effect of lattice heating on T_c and in this experiment, the effect will be a few tenmillidegrees.

One of the most interesting results shown in Fig. 3 is that R larger than Rn is observed in the temperature range very near T_c. The photoinduced resistance is not observed in normal state so that the heating of lattice will not produce the large resistance. This result suggests that the anomolous large resistance would be mainly due to the effect of photoexcited quasiparticles.

The $I-V$ characteristics of the resistive state as a function of reduced temperature, $t=T/T_c$, is shown in Fig. 4. The $I-V$ characteristics show the linear relation

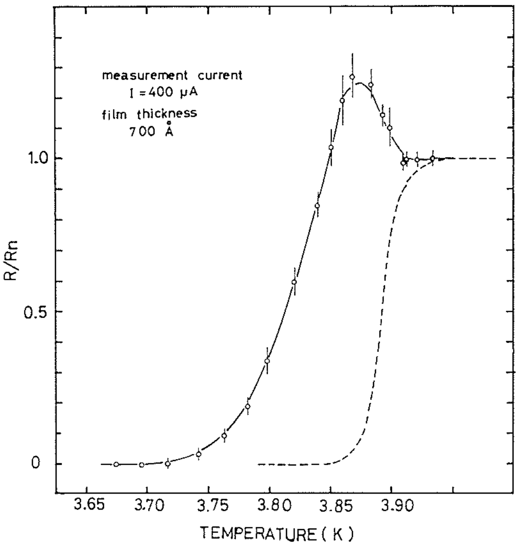

Fig. 3. R/Rn versus temperature. Transition curve in zero light is shown by the dashed line.

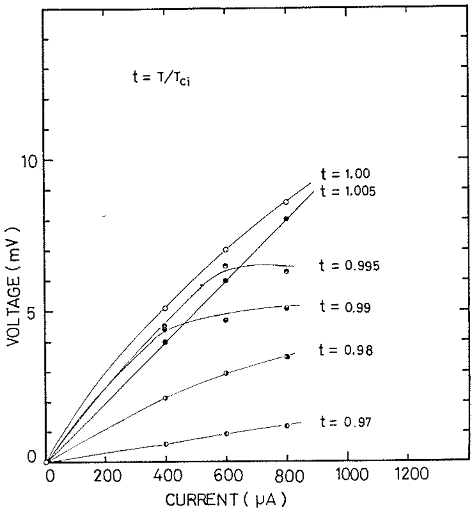

Fig. 4. The current-voltage characteristics as a function of reduced temperature, $t(=T/T_c)$. T_c is the superconducting transition temperature determined in zero light.

in the range of temperature above $t=1.005$, i. e., where Ohm's law is satisfied. While in the temperature range below $t=1.00$ the $I-V$ characteristics show the nonlinear relations, in which current increases rapidly with the increasing voltage. These characteristics can not be explained in terms of a thermal equilibrium or T^* model[6]. Recently several workers[7,11] pointed out that spatially inhomogeneous state would be established in externally excited superconducting films. For the initially induced inhomogeneous state, it may be due to the nonuniform illumination, spatial variation of fluctuations and/or phase slip occurs in flow of current[12,13]. These spatial inhomogeneity would result in local concentration of the excess quasiparticles and phonons and these regions may be driven into normal state. For nonlinear $I-V$ characteristics, one possibility is that as the current increases injection of superelectrons through the normal-superconducting interface increases because of the gradient of chemical potential. Therefore it is likely that spatially inhomogeneous state and its instability are dominant for the nonequilibrium state near T_c.

4.1 Relaxation Time

Relaxation times of the resistance induced by laser illumination, τ_{relax}, as a function of Δ_{Sn}/k_BT are shown in Fig. 5, where Δ_{Sn} is the superconducting energy gap calculated from BCS formula in tin, and T is an initial temperature before laser light illumination. In this temperature region, the relaxation time decreases rapidly with the decreasing temperature.

In this experiment, an obtained relaxation time involves both an excited quasiparticle relaxation time, $\tau_{\text{q.relax}}$, in which the quasiparticle recombines with another quasiparticle again and a thermal relaxation time, τ_{thermal}, in which the heated films relax toward its equilibrium temperature. Therefore our obtained relaxation times are approximately represented by the equation

$$\tau_{\text{relax}}^{-1} = \tau_{q.\,\text{relax}}^{-1} + \tau_{\text{thermal}}^{-1}\,. \tag{1}$$

In this system, thermal relaxation times are mainly determined by the thermal contact between the Sn films and the substrate; (1) According to the experiments of heat pulse transmission by Gutfeld[14], thermal relaxation time is estimated to be 10^{-8} sec which is shorter by one or two orders compared with our relaxation time. (2) As discussed in Sec. 3 the lattice heating is estimated to be small in the photoinduced resistance. It is thus concluded that the obtained relaxation times are almost reflected by the excited quasiparticle relaxation.

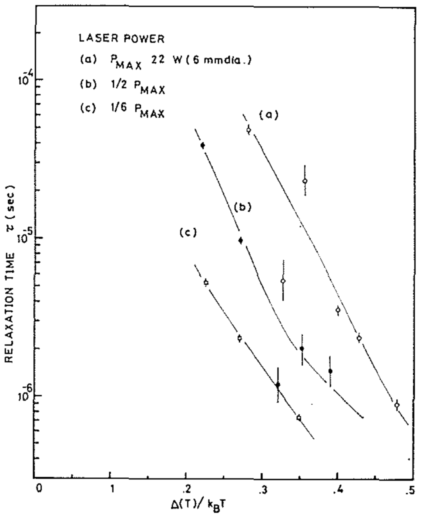

Fig. 5. Relaxation times of photoinduced resistive state as a function of $\Delta/k_B T$. Δ is the energy gap without illumination. Maximum laser power is 22 W (6 mmdia.) and film thickness 700 Å.

In the quasiparticle relaxation we must consider two steps; First is a process in which the excited quasiparticles thermalize with respect to the lattice and then the relaxation process which produces Cooper pairs due to a recombination. At low temperatures, where $T \ll T_c$, the recombination process is explained by the bottle-neck model.[5] The state density of quasiparticle at the energy gap decreases with the increasing temperature and energy levels which will be possible to recombine broaden compared with those at low temperatures. Therefore, upon approaching the transition temperature the bottle-neck model can not be applied for the relaxation process.

The rate equations of Rothwarf and Taylor[8] that describe the nonequilibrium quasiparticle and phonon densities in a superconducting film are

$$\frac{\partial N}{\partial t} = I_0 + \beta N_\omega - RN^2\,, \tag{2}$$

$$\frac{\partial N_\omega}{\partial t} = \frac{1}{2} RN^2 - \frac{1}{2}\beta N_\omega - \tau_\gamma^{-1}\big(N_\omega - N_\omega(T)\big)\,, \tag{3}$$

where N is the number density of quasiparticles, I_0 the volume rate of creation of quasiparticles per second by an external mechanism, N_ω the number density of phonons with energy greater than 2Δ, β the transition probability for breaking of paris by phonons, R a recombination coefficient, τ_γ^{-1} the net transition probability for loss of phonons from an energy range greater than 2Δ, and $N_\omega(T)$ the thermal equilibrium number of phonons with energy $> 2\Delta$. Excess quasiparticle number density, ΔN, in the steady state is

$$\Delta N = N(T)\left(\left(1 + \frac{I_0}{N(T)}\,\tau_{eff}\right)^{1/2} - 1\right)\,, \tag{4}$$

where

$$\tau_{eff} = \frac{1}{RN(T)} + \frac{\beta\tau_r}{2RN(T)}, \tag{5}$$

and $N(T)$ is the thermal equilibrium number of quasiparticles.

We put $\tau_R = (RN(T))^{-1}$ and $\tau_{pq} = (\beta/2)^{-1}$ which are the intrinsic recombination time and the mean lifetime of phonons for creating quasiparticles. Then the effective quasiparticle lifetime is

$$\tau_{eff} = \tau_R(1 + \tau_r/\tau_{pq}). \tag{6}$$

Under the condition of $\tau_r \ll \tau_{pq}$, the phonons are almost in thermal equilibrium and the effective quasiparticle lifetime is almost that of the intrinsic recombination time. Within the limit of $\tau_r \gg \tau_{pq}$, a recombination phonon is likely to be reabsorbed and to create two quasiparticles rather than to escape from the superconductor. Then the distribution of phonon is far from equilibrium and the quasiparticle lifetime strongly depends on the number of high energy phonons.

4.2 Recombination Rate Near T_c

Detailed calculations of the quasiparticle recombination rate due to phonon emission at low reduced temperature have been given by Rothwarf and Cohen[15] on the assumption of a spherical Fermi surface. In a process in which a created quasiparticle (momentum and spin $\boldsymbol{k}\uparrow$) combines with another quasiparticle ($-\boldsymbol{k}'\downarrow$) to form a Cooper pair with emission of a phonon, the phonon of momentum $\boldsymbol{q} = \boldsymbol{k}\uparrow - \boldsymbol{k}'\downarrow + \boldsymbol{K}$. The vector $\boldsymbol{K}$ is any reciprocal lattice vector and we assume that $\boldsymbol{K} = 0$ (normal process). Then the recombination rate is

$$W = \frac{A}{(2\pi)^2\hbar} \int_{k'>k_F} \left(1 + \frac{\Delta^2}{EE'}\right) \frac{|\boldsymbol{k}-\boldsymbol{k}'|\,\delta(E+E-\hbar s_L|\boldsymbol{k}-\boldsymbol{k}'|)}{1+\exp(E'/k_BT)}\, d^3k', \tag{7}$$

where

$$A = (\hbar N/2Ms_L)(4\pi Ze^2/k_s^2)^2, \qquad E = (\varepsilon^2 + \Delta^2)^{1/2},$$

s_L is the longitudinal sound velocity, k_F the Fermi momentum, Z the number of conduction electrons per ion, ε the energy measured relative to the Fermi energy, and k_s the inverse Fermi-Thomas screening length. E and E' are energes corresponding to $\boldsymbol{k}$ and $\boldsymbol{k}'$. Approximating $|\boldsymbol{k}-\boldsymbol{k}'|$ by $2k_F \sin(\theta/2)$ and integrating over angles removes the delta function. As $E_F \gg \hbar\omega_D$, we obtain

$$W_N = \frac{2\pi}{9}\,\frac{\hbar E_F^2}{(\hbar s_L)^4 \rho_m k_F^2} \int_\Delta^\infty \left(1 + \frac{\Delta^2(\mathrm{T})}{EE'}\right)(E+E')^2 N(E') f(E')\, dE' \tag{8}$$

where

$$N(E') = N(0)\frac{E'}{\sqrt{E'^2 - \Delta^2(T)}},$$

$f(E')$ is a quasiparticle Fermi distribution function, ρ_m the mass density, $N(0)$ the density of state at the Fermi energy in the normal state and ω_D the Debye frequency of phonon.

At low temperatures, the energy range which the probability of the quasiparticle recombination is larger than that of relaxation becomes narrow near the energy gap, because of there being a large state density of quasiparticle. While near T_c, the energy range broadens and the thermalization time is longer than the intrinsic recombination time. The average energy of quasiparticles is determined by the thermal energy rather than the energy gap. This is shown in Fig. 6 and the average quasiparticle energy is

$$\bar{E}=\int_{\varDelta}^{\infty} E'N(E')f(E')\,dE' \Big/ \int_{\varDelta}^{\infty} N(E')f(E)\,dE' \,. \qquad (9)$$

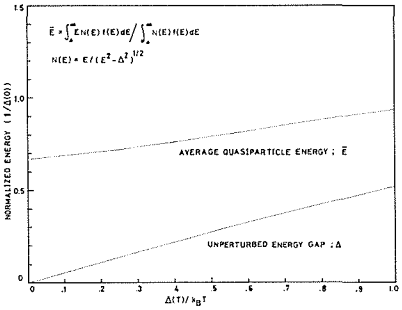

Fig. 6. The average quasiparticle energy and the energy gap in an equilibrium superconductor. Near T_c, at these energy levels a quasiparticle recombines with another quasiparticle before attaining equilibrium.

Near T_c a quasiparticle created at a level of high energy will recombine with another quasiparticle before attaining thermal equilibrium. For Sn, taking that $\rho_m=7.31\,(\mathrm{g/cm^3})$, $E_F=4.4\,(\mathrm{eV})$, and $s_L=3.32\times 10^5\,(\mathrm{cm/sec})$, as a quasiparticle recombines at the energies, the energy gap and the average quasiparticle energy respectively, a recombination coefficient R can be calculated from eq. (8) (This is shown in Fig. 7.). For the energies, the intrinsic recombination time τ_R is shown in Fig. 8. For the energies, recombination probability decreases with the increasing temperature. These τ_R are rather shorter than the experimental value shown in Fig. 5. Therefore as discussed in Sec. 4. 1, high energy phonons will strongly affect the quasiparticle recombination process and the system will be under the condition where $\tau_\gamma \gg \tau_{pq}$. Within this limit, from eq. (6), an effective recombination time is given by $\tau_{eff}=\tau_R\tau_\gamma/\tau_{pq}$. A graph of the relation between τ_γ/τ_{pq} and the normalized energy gap $\varDelta/k_BT$ is shown in Fig. 9, assuming that a quasiparticle recombines at each energy, (a) $\varDelta$ and (b) $\bar{E}$. As the ambient temperature decreases, the rate of absorption of phonons by pairs decreases and the probability of which the phonon escapes from film increases. It has been estimated in superconducting films that $\tau_\gamma=4d/\eta c_s$ under the condition where the phonons scatter diffusely at the film surface and leave the film only at the interface between the film and substrate. Where η is the average phonon transmission probability at superconductor surface, c_s the sound velocity and d the film thickness. Taking that $\eta=0.08$[1,17] (from the acoustic mismatch the-

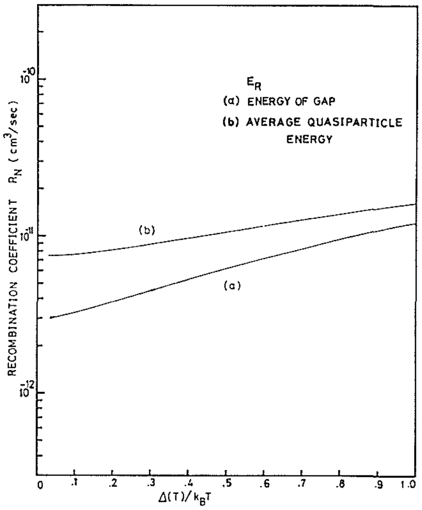

Fig. 7. Recombination coefficient R_N in normal process as a function of $\varDelta/k_BT$. Quasiparticle recombines with another quasiparticle at the energies (E_R); (a) the energy corresponding to $\varDelta$ and (b) the average quasiparticle energy.

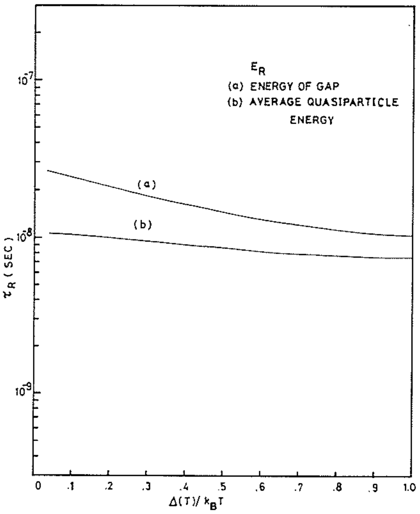

Fig. 8. Intrinsic recombination time τ_R which is calculated from eq. (8) for the same case in Fig. 7.

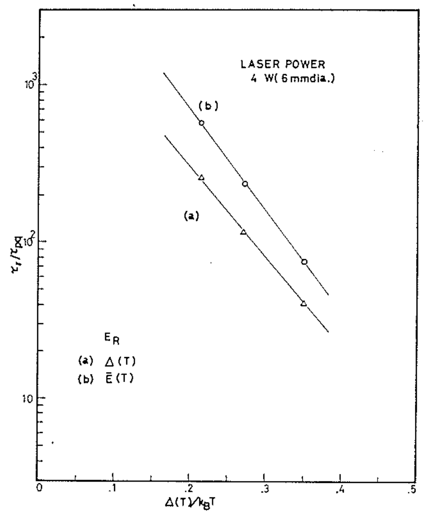

Fig. 9. τ_γ/τ_{pq} versus $\Delta(T)/k_BT$ for the same case in Fig. 7. $\Delta(T)$ is determined without illumination.

ory), we calculate that τ_γ is of order of 10^{-9} sec and has a weak temperature dependence. It is thus likely that τ_{pq} is dominant our experiment.

Comparing these relaxation times with the experimental value, we see that near T_c the interaction between quasiparticle and phonon plays an important role for the establishment of equilibrium in this system. When the ambient temperature is low, the increase of the pair-breaking time τ_{pq} arising from collisions between phonons and pairs would occur by a decrease of the number density of phonons which are effective for the pair breaking. Therefore, it is concluded that the condensation energy of quasiparticle to form pairs becomes large with decreasing of temperature in the photoinduced resistive state, while by the T^* model the condensation energy is almost constant over the above temperature range.

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