Transient responses of superconducting lead films measured with picosecond laser pulses

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(Received 25 July 1980)

Picosecond laser pulses are used, for the first time, to excite superconducting Pb films out of equilibrium. Pb microbridges are used to measure the threshold power needed to drive the Pb film normal as a function of temperature. Pb tunnel junctions are used to measure the quasiparticle relaxation times at temperatures close to T_c and at low reduced temperatures. The measured quasiparticle relaxation time for a 500-Å Pb$-$PbO$_x$$-$500-Å Pb tunnel junction is 3.1 nsec for the sample in vacuum at temperatures near T_c, and 2.5 nsec for the sample in superfluid at low reduced temperatures. At temperatures near T_c, the quasiparticle relaxation time for Pb is shown to be identical to the phonon escape time instead of the effective quasiparticle recombination time. At low reduced temperatures, the quasiparticle relaxation, for the case of excess quasiparticle density being much larger than the thermal equilibrium value, is shown to be approximately exponential with a time constant equal to twice the phonon escape time. The phonon transmissivities for the Pb-quartz, Pb-superfluid He, and Pb-PbO$_x$ interfaces are determined from these relaxation time measurements to be 0.16, 0.16, and 0.2, respectively.

I. INTRODUCTION

Relaxation-time measurements of various superconducting materials have been a principal subject in the general field of nonequilibrium superconductivity.[1,2] Laser light has frequently been used to perturb the superconductors for studying either the steady-state response[3–10] or the transient behavior.[11–17] Most of the transient studies were done for superconductors with weak electron-phonon coupling, such as Al and Sn. In this paper, an experimental study of the transient responses of superconducting Pb films is presented. It is the first time that the response of such films to picosecond laser pulses (20 to 50 psec) has been reported. Previously, superconducting Pb films irradiated with microsecond laser pulses have been studied by Testardi[11] and Golovashkin *et al.*,[17] and superconducting bulk Pb samples irradiated with nanosecond laser pulses have been studied by Hu *et al.*[15]

It is well known that a strong electron-phonon coupling material has a very short characteristic electron-phonon interaction time.[18] For example, Pb at its superconducting transition temperature T_c has an inelastic scattering time for electrons at the Fermi energy of about 10 psec, and the lifetime of phonons with energy $\sim k_B T_c$ of about 60 psec.[18] Since the total energy of electrons and phonons is conserved in the electron-phonon interaction process, a characteristic energy releasing time of the metal film is also very important to the transient behavior. For thin films on dielectric substrates subject to a spatially uniform excitation, the characteristic energy releasing time is the phonon escape time,[19] τ_γ, into the substrate or into the He bath if the thin films are in direct contact with He. It is intuitively clear that τ_γ can be roughly estimated to be the phonon transit time across the film multiplied by the inverse of the phonon transmission probability (phonon transmissivity) of the interface. For a 1000-Å Pb film, τ_γ is at least 0.2 nsec. Therefore, with a laser pulse width comparable to the electron-phonon interaction times and much shorter than the phonon escape time τ_γ, we have the advantage of making the following studies, which cannot be made with longer laser pulses. First, we studied the threshold power, or more precisely the threshold energy per pulse, needed to drive the superconducting film into the normal state. It is clear that if the laser pulse is comparable to or longer than τ_γ, the threshold power would be τ_γ dependent instead of intrinsic. Second, from the relaxation-time measurements of the superconducting Pb tunnel junctions, we obtained the information on τ_γ's for Pb-quartz, Pb-superfluid He, and Pb-PbO$_x$ interfaces. These τ_γ's turn out to be roughly ten times longer than the phonon transit time across the film, i.e., of the order of several nanoseconds. We believe that this is the first time the direct measurement of phonon escape times for superconducting Pb films has been reported.

The sample fabrication and the experimental setups are briefly described in the next section. Experimental results are presented together with the theoretical interpretations in the third section. Comparison with other available information on the threshold power and τ_γ's can be found in the final section with a brief discussion of the implications of the present experiment.

II. EXPERIMENT

The samples used in this experiment were either variable-thickness microbridges of superconducting Pb or Pb-PbO_x-Pb tunnel junctions. *C*-axis crystalline quartz disks were always used as substrates. These were 250 μm thick and 1 in. in diameter. Pb was deposited onto the substrates by conventional hot-boat evaporation in 10^{-5} to 10^{-6} torr vacuum. The geometry and the dimension of Pb microbridges is shown in the inset of Fig. 1. The Pb tunnel junctions were cross-strip type with a tunneling area about 75×100 μm^2. The thickness of the individual films of the tunnel junctions varied from 500 to 4000 Å. Both equal-thickness and unequal-thickness cases have been investigated experimentally. The normal-state resistances of our tunnel junctions were typically 1 Ω. We only present data for high quality tunnel junctions which show negligible leakage current for $V < 2\Delta/e$ at low reduced temperatures. The film thicknesses and the normal-state resistances for those samples are listed in Table I.

Samples were either mounted in an optical cold-finger Dewar for experiments done in the temperature range of 5 to 7 K, or mounted in an optical immersion Dewar for experiments done in the temperature range of 1.5 to 2 K. In the former case the samples were in vacuum, and in the latter case the samples were in direct contact with superfluid He.

Short laser pulses produced by a dye laser synchronously pumped by a mode-locked Ar laser were used to irradiate the superconducting samples. The laser pulse width was typically 20 to 50 psec. The exact pulse width is not important in the present experiment as long as it is much shorter than the phonon escape time τ_γ. The time interval between laser pulses was fixed at 14 nsec.

Superconducting microbridges were constant-current biased at a current level much less than the critical current of the bridge. Because a superconducting microbridge is essentially an ideal on-off switch, it serves as a threshold detector to detect the minimum light power needed to drive the superconducting Pb film normal. When the light power just exceeds the threshold power, both the rise time and the fall time of the voltage pulses of the microbridges are essentially limited by our detection resolution; this feature was actually used to check our overall detection resolution experimentally. Superconducting tunnel junctions were usually biased at $V = 2\Delta/e$, where the dynamic resistance is close to the minimum. The *RC* time constant at the bias point is at least ten times smaller than the *RC* time constant at $V > 2\Delta/e$, which is estimated to be 0.3 nsec. The voltage pulses, which reflect the changes in the superconducting gaps, were transmitted through a coaxial cable, and amplified by two fast amplifiers (B&H DC3002, rise time ~130 psec) in tandem, and finally recorded by a transient digitizer (Tektronix 7912 AD) with less than 1-nsec rise time. Our overall detection resolution time is both estimated and determined to be about 1 nsec.

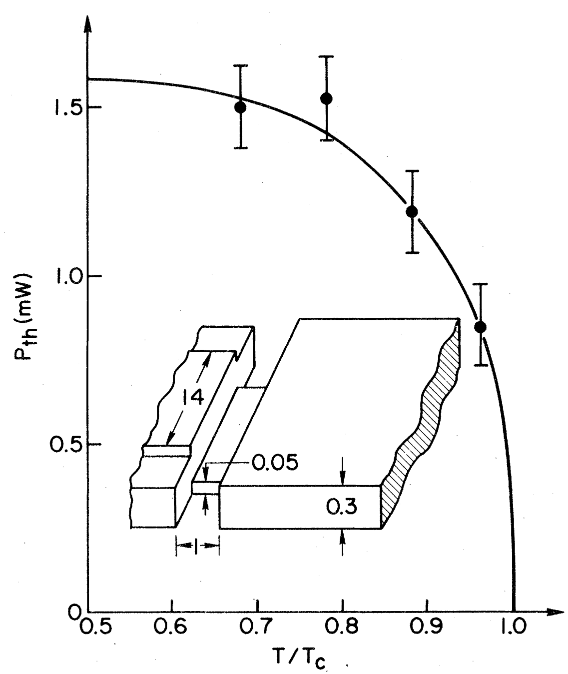

FIG. 1. Measured average threshold power of the laser pulse vs reduced temperature. The sample geometry is schematically shown in the inset with dimensions indicated in the units of μm. Our laser pulse has a pulse width of approximately 30 psec and a pulse period of 14 nsec. The laser peak power and the energy per pulse can be obtained by multiplying the measured average power by 470 and 1.4×10^{-8} sec, respectively.

TABLE I. A list of the film thicknesses of each film, normal-state resistance R_N, and the experimental temperature range for the samples tested and analyzed.

Sample No.	Film 1 (top) (Å)	Film 2 (bottom) (Å)	R_N (Ω)	T/T_c
1	500	500	1.0	0.7 ~ 0.9
2	500	1500	0.8	0.7 ~ 0.9
3	4000	4000	1.2	0.7 ~ 0.9
4	1000	1000	3.8	0.7 ~ 0.9
5	500	2500	1.3	0.7 ~ 0.9
6	500	500	4.0	0.2 ~ 0.3
7	500	2500	0.5	0.2 ~ 0.3
8	500	500	20.0	0.2 ~ 0.3

III. EXPERIMENTAL RESULTS AND INTERPRETATIONS

A. Microbridges

Because of the fact that the microbridges are ideal on-off switches, the shape of the voltage pulses induced by laser irradiation depends on the laser power and is, in general, nonexponential. We do not intend to present the analysis of the pulse shapes here. Instead, the measured threshold power as a function of reduced temperature is shown in Fig. 1. The threshold power is defined to be the minimum peak power of the laser pulse needed to drive the superconducting film normal with zero current bias. In practice, some small current bias is needed to give us observable signals, and the zero-current limit is obtained by extrapolation of the measurements of threshold power versus bias current at a fixed temperature. The solid line in Fig. 1 is a theoretical fit with the absolute absorbed energy per pulse as the fitting parameter. The theoretical calculation of the threshold power as a function of reduced temperature was carried out using the Rothwarf-Taylor equations[19] for quasiparticles and 2Δ phonons, i.e., phonons with energy greater than or equal to twice the energy gap parameter Δ, as follows:

$$\dot{N}_q = I_q - RN_q^2 + 2\tau_B^{-1}N_p \ , \tag{1}$$

$$\dot{N}_p = I_p + \tfrac{1}{2}RN_q^2 - \tau_B^{-1}N_p - \tau_\gamma^{-1}(N_p - N_{pT}) \ , \tag{2}$$

where N_q, N_p are the number density of quasiparticles and 2Δ phonons, respectively, N_{qT} and N_{pT} are the thermal equilibrium values of N_q and N_p at temperature T, I_q and I_p are the external generation rates, R is the rate for one quasiparticle to recombine with any other quasiparticle, τ_B and τ_γ are the phonon pair breaking time and phonon escape time, respectively. The quadratic terms in N_q reflect the fact that it takes two quasiparticles to recombine to make one pair. It is true that the Rothwarf-Taylor (RT) equations are crude approximations to the complicated quasiparticle and phonon collision integrals. However, there is theoretical[20] and experimental[1] evidence to show that the use of the RT equations can give results reasonably close to the more elaborate collision-integral calculations. Because the cooling processes of a high-energy quasiparticle are not included in the RT equations, the quasiparticles and phonons generated by cascading a direct photon-excited quasiparticle from high energy (~ 1 eV) to low energy (between Δ and 2Δ) have to be included in the generation terms I_q and I_p. The estimated cascading time is in the order of 1 psec for superconducting Pb.[21] Therefore we expect the RT equations to give a reasonable account of the quasiparticle and phonon system on a time scale longer than 1 psec. The same estimate gives the ratio of I_q to I_p for 2-eV-photon pumping to be 1:15. In addition, for superconducting Pb, the following values are assigned for the parameters used in the RT equations[20]:

$$4N(0)\Delta_0 R = 2\times 10^{12}\ \text{sec}^{-1} \ ,$$

$$\tau_B = 3\times 10^{-11}\ \text{sec} \ ,$$

where $N(0) = 2.1\times 10^{19}$ (meV)$^{-1}$ cm^{-3} is the single-spin electron density of states at the Fermi energy, and $\Delta_0 = 1.4$ meV is the zero-temperature superconducting gap of Pb. For the calculation of the threshold power, both I_q and I_p are assumed to be Gaussian in time with a full width at the half maximum of about 30 psec. The peak values of N_q and N_p are numerically determined from the Eqs. (1) and (2). It has been verified numerically that these are very insensitive to the value of τ_γ, provided that τ_γ is much longer than 30 psec. From the numerical calculation, the peak value of N_q can be determined as a function of the peak value of I_q (the ratio of I_p/I_q is kept constant at 15). We assume that the nonequilibrium quasiparticle distribution due to laser excitation is close to the T^* model[12,22]; hence the superconductor is driven normal when $N_q/4N(0)\Delta$ reaches 0.4.[12] The threshold power, P_{th}, is calculated from the corresponding I_q and I_p by using the following formula:

$$P_{\text{th}} = I_q\Delta(T) + I_p 2\Delta(T) = 31 I_q \Delta(T) \ , \tag{3}$$

where $\Delta(T)$ is the superconducting gap at T.

There is a large uncertainty in determining the absolute energy per pulse absorbed by the microbridge from the uncertainty in estimating the laser spot size. Thus the absolute scale of the threshold power is used as an adjustable parameter to fit the experimental data. The fitting is satisfactory. The threshold power per unit area determined from the experimental estimates and the theoretical calculation agree with each other to within the experimental uncertainty. The experimental estimates give a value between 3 and 30 kW/cm^2, which corresponds to between 20 and 200 mJ/cm^3 per pulse for a 500-Å Pb film, at low reduced temperatures.

B. Tunnel junctions

1. Theory

In thermal equilibrium, the superconducting gap Δ at finite temperature T is related to the thermal distribution function of the quasiparticles,

$$f_T(E) \equiv [1+\exp(E/k_B T)]^{-1} \ ,$$

through the BCS gap equation:

$$\Delta/\Delta_0 = \exp\left(-\int_{-\infty}^{\infty} d\epsilon\, f_T(E)/E\right) \ , \tag{4}$$

where $\epsilon \equiv \pm(E^2-\Delta^2)^{1/2}$.

It has been shown[23] that the BCS gap equation is still valid for superconductors subject to a constant external perturbation if the nonequilibrium gap and quasiparticle distribution function are used in Eq. (4). In a non-steady-state situation, there are reasons[24] to believe that Eq. (4) still holds instantaneously provided that the relative rate of change in Δ, i.e., $(\dot{\Delta}/\Delta_0)$, predicted by Eq. (4) through the change in quasiparticle distribution, is smaller than $\Delta/\hbar$. The relaxation of quasiparticles cannot be faster than the quasiparticle recombination time (about 3 psec for the thermally excited quasiparticles in Pb) near T_c. That implies that Eq. (4) is instantaneously valid unless $T/T_c \gtrsim 0.999$.

It has also been shown[22] that the superconducting gap determined from Eq. (4) is predominantly determined by the quasiparticle density rather than the details of the distribution function so that Eq. (4) can be approximated by

$$\Delta/\Delta_0 = 1 - 2N_q/4N(0)\Delta_0 \ . \tag{5}$$

Within the approximation made for Eq. (5), which is valid for our experiment, the change in superconducting gap measured experimentally is a direct measurement of the changes in quasiparticle density N_q.

In order to understand both the dominant quasiparticle relaxation time and the pulse shape and their temperature dependences, it is very helpful to model the tunnel junction within the framework of the RT equations [Eqs. (1) and (2)].

First of all, to understand the difference in quasiparticle relaxations at temperatures close to T_c and low reduced temperatures, let us consider a simpler situation: a single film with only one interface transmitting phonons with a phonon transmissivity η. The phonon escape time τ_γ can be calculated from

$$\tau_\gamma = 4d/\eta v_s \ , \tag{6}$$

where d is the film thickness and v_s is the phonon velocity.

At temperatures close to T_c, the thermal population of quasiparticles is large [0.1 to 0.3 in units of $4N(0)\Delta_0$] compared with the excess quasiparticle population (of the order of 0.02) generated by laser pulses to produce a small but detectable signal, say of the order of 40 μV.[25] Therefore Eqs. (1) and (2) can be linearized into

$$\Delta\dot{N}_q = I_q - \tau_R^{-1}\Delta N_q + 2\tau_B^{-1}\Delta N_p \ , \tag{7}$$

$$\Delta\dot{N}_p = I_p + \tfrac{1}{2}\tau_R^{-1}\Delta N_q - \tau_B^{-1}\Delta N_p - \tau_\gamma^{-1}\Delta N_p \ , \tag{8}$$

where $\Delta N_q \equiv N_q - N_{qT}$, $\Delta N_q \equiv N_p - N_{pT}$, and $\tau_R^{-1} \equiv 2N_{qT}R$.

The solution of Eqs. (7) and (8) can be written as

$$\Delta N_q(t) = Ae^{-t/\tau_+} + Be^{-t/\tau_-} \ , \tag{9}$$

$$\Delta N_p(t) = -A\,\alpha_+ e^{-t/\tau_+} - B\,\alpha_- e^{-t/\tau_-} \ , \tag{10}$$

where

$$\tau_\pm^{-1} \equiv \tfrac{1}{2}\{\tau_R^{-1} + \tau_B^{-1} + \tau_\gamma^{-1} \pm [(\tau_R^{-1} + \tau_B^{-1} + \tau_\gamma^{-1})^2 - 4\tau_R^{-1}\tau_\gamma^{-1}]^{1/2}\}$$

and

$$\alpha_\pm \equiv (\tau_\pm^{-1} - \tau_R^{-1})/2\tau_B^{-1} \ .$$

For superconducting Pb near T_c, we have $\tau_R^{-1} > \tau_B^{-1} >> \tau_\gamma^{-1}$. The two relaxation times $\tau_\pm$ of the quasiparticle-phonon system reduce to

$$\tau_\pm^{-1} \sim \begin{cases} \tau_R^{-1} + \tau_B^{-1} \\ \tau_\gamma^{-1} \end{cases} .$$

Since our experimental detection resolution time is longer than τ_+, it is clear that the measured quasiparticle relaxation time is exactly the phonon escape time τ_γ.

On the other hand, if τ_R is the longest time among the three time constants in Eqs. (7) and (8), which is appropriate for superconducting Al for example, we have

$$\tau_\pm^{-1} \sim \begin{cases} \tau_B^{-1} + \tau_\gamma^{-1} \\ \tau_R^{-1}/(1 + \tau_\gamma/\tau_B) \end{cases} .$$

In this case, the dominant quasiparticle relaxation time is the effective quasiparticle recombination time, $\tau_{\text{eff}} \equiv \tau_R(1 + \tau_\gamma/\tau_B)$. It is interesting to note that the steady-state solution of ΔN_q from Eqs. (7) and (8) for a constant quasiparticle injection rate I_q is always given by $I_q\tau_{\text{eff}}$ irrespective of the relative magnitudes of τ_R, τ_B, and τ_γ.[19]

At low reduced temperatures, say $T/T_c \lesssim 0.3$, the thermal population of quasiparticles is extremely small $[\lesssim (10^{-3})4N(0)\Delta_0]$. In order to produce an experimentally observable signal, the excess quasiparticle density created by an external perturbation has to be much larger than the thermal value. Therefore the RT equations cannot be linearized and their solutions are no longer simple exponential functions in general. However, for the case of Pb, the quasiparticle recombination rate constant R in Eqs. (1) and (2) is so large that the condition $\tau_\gamma^{-1} << RN_q$ is still valid before N_q decays to some undetectably small value. Approximate solutions of N_q and N_p for the nonlinear RT equations [Eqs. (1) and (2)] can be found as follows:

$$N_q(t) = N_{q0}e^{-t/2\tau_\gamma} - N_{q0}/4\tau_\gamma RN_{q0} \ , \tag{11}$$

$$N_p(t) = N_{p0}e^{-t/\tau_\gamma} \ , \tag{12}$$

where the initial values of N_{q0} and N_{p0} are related by

$$2N_{p0}/N_{q0}^2 = \tau_B R \ . \tag{13}$$

This is a result of a self-consistent assumption that on a time scale shorter than τ_γ a quasiequilibrium between the quasiparticles and phonons is reached due to their fast interaction rates. The validity for the self-consistency requires that $4\tau_\gamma R N_{q0} >> 1$. For any small detectable signal ($\gtrsim 40\ \mu$V), we estimate that $4RN_{q0} \gtrsim 10^{11}\ \text{sec}^{-1}$. So the requirement for this approximation to be valid is fully satisfied with τ_γ expected to be in the order of nanoseconds.

The interesting fact which comes out of this analysis is that the excess quasiparticle density still decays approximately exponentially at low reduced temperatures but the time constant changes from τ_γ to $2\tau_\gamma$.[26]

To describe a tunnel junction, which was actually used to measure the quasiparticle relaxation experimentally, we need four coupled equations and three phonon transmissivities; this system of equations is too complicated to solve without making any approximations. Fortunately, based upon the fact that our signal is only proportional to the excess quasiparticle density and that this excess quasiparticle density decays exponentially either with a time constant τ_γ or $2\tau_\gamma$ depending on whether the experiment was carried out near T_c or at low reduced temperatures, we can simplify the four coupled equations into two as follows:

$$\dot{N}_1 = -\chi_1(N_1 - N_2) - \gamma_1(N_1 - N_{1T}) \ , \tag{14}$$

$$\dot{N}_2 = -\chi_2(N_2 - N_1) - \gamma_2(N_2 - N_{2T}) \ , \tag{15}$$

where N_1 and N_2 represent the quasiparticle densities in the film 1 and 2, respectively, χ_1^{-1} (χ_2^{-1}) and γ_1^{-1} (γ_2^{-1}) are simply the phonon escape times or twice as much depending on the temperature from the film 1 (2) into film 2 (1) and through the other interface into the thermal bath, respectively. The phonons do not enter into these two equations explicitly, but they implicitly control the quasiparticle decay rates through χ's and γ's in the Eqs. (14) and (15).

Experimentally, our signals are proportional to the changes of the sum of the superconducting gaps of the two films, which are, in turn, proportional to the change of the sum of the quasiparticle densities of the two films through Eq. (5). It is convenient to rearrange Eqs. (14) and (15) into the following forms:

$$\dot{N}_s = -(\gamma_s/2)N_s - (\chi_d + \gamma_d/2)N_d \ , \tag{16}$$

$$\dot{N}_d = (\gamma_d/2)N_s - (\chi_s + \gamma_s/2)N_d \ , \tag{17}$$

where

$$N_s, \chi_s, \gamma_s \equiv N_1 + N_2, \chi_1 + \chi_2, \gamma_1 + \gamma_2$$

and

$$N_d, \chi_d, \gamma_d \equiv N_1 - N_2, \chi_1 - \chi_2, \gamma_1 - \gamma_2 \ .$$

Since the thicknesses of the Pb films used in our experiment are always larger than the penetration depth of the light, only one of the two films was directly excited by the laser pulse, say film 1. The following initial condition has to be imposed on the solutions of Eqs. (16) and (17):

$$N_s(t=0) = N_d(t=0) = N_1(t=0) \ .$$

Then the solution of $N_s(t)$ can be written as

$$N_s(t) = Ae^{-tR_+} + Be^{-tR_-} \ , \tag{18}$$

where

$$R_\pm \equiv \tfrac{1}{2}\{(\chi_s + \gamma_s) \pm [(\chi_s + \gamma_s)^2 - 4(\gamma_1\gamma_2 + \gamma_1\chi_2 + \gamma_2\chi_1)]^{1/2}\} \ , \tag{19}$$

$$A \equiv (\chi_d + \gamma_1 - R_-)/(R_+ - R_-) \ , \tag{20}$$

$$B \equiv (R_+ - \chi_d - \gamma_1)/(R_+ - R_-) \ . \tag{21}$$

For convenience of writing and discussion, the constant $N_1(t=0)$ which should appear as a multiplicative factor in Eqs. (20) and (21), has been dropped. It is true that R_- is always less than R_+, but it does not necessarily imply that R_-^{-1} is the dominant relaxation time for our signals. Because all the χ's and γ's are phonon escape rates of the same order of magnitudes, the constants A and B in Eq. (18) have to be evaluated for individual experimental situations.

2. *Experimental results*

Figure 2 shows a typical junction I-V curve and the schematics of the junction geometry. The typical normal-state resistance R_N of our samples is 1 Ω. The capacitance of our junctions ($17 \times 100\ \mu\text{m}^2$) is estimated to be 300 pF. Since we bias the junction at the current rising portion of $V = 2\Delta/e$, the dynamic RC time constant is expected to be much less than $R_N C \approx 300$ psec. In order to make sure no serious error was made in the estimate of junction capacitance, a junction with normal-state resistance of about 20 Ω was made and tested. The rise time and the relaxation tail of the signal are comparable to the typical signals for junctions of much smaller normal-state resistance.

Table I summarizes the sample number, the film thickness of each film of the junction, the junction normal-state resistance, and the experimental temperature range. For convenience of discussion, the film on top is named film 1 and the film in direct contact with the substrate, film 2. A thin 30-Å PbO_x layer separates film 1 from film 2; the other surface of film 1 is either in vacuum for the experiments conducted at the higher temperatures ($0.7 \lesssim T/T_c \lesssim 0.9$) or in contact with superfluid He for the experiments conducted at the lower temperatures.

In both temperature ranges, the smallest signal we measured and also used to analyze the relaxation

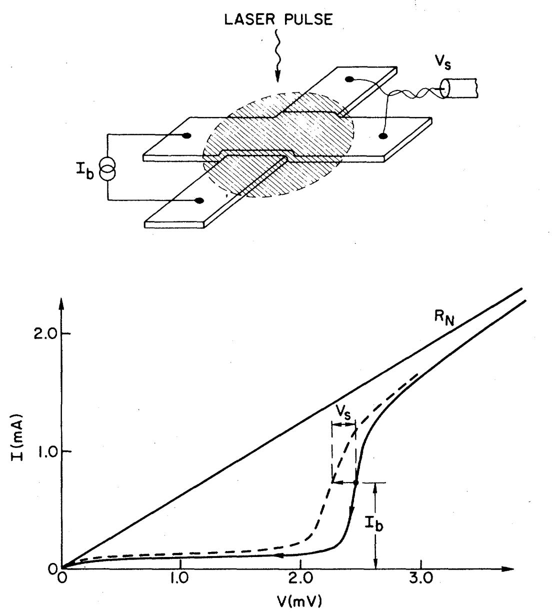

FIG. 2. Typical I-V curve (solid line) for our Pb-PbO$_x$-Pb tunnel junctions at $T/T_c \approx 0.7$ and a schematic of the sample geometry and the measurement. The dashed line indicates the laser-excited, transient I-V curve. V_s is the observed voltage pulse which has an opposite polarity with respect to the bias current I_b. Since the transient I-V curve cannot exceed the normal state I-V (shown as the straight line with a slope R_N) there is a maximal voltage signal $V_s \approx \Delta(T)/e \approx 1$ mV, with the current bias I_b as shown.

time is about 40 μV. Since the junction bias point is approximately in the middle of the current rising portion of $V \approx 2\Delta/e$, the maximum signal amplitude is roughly equal to Δ ($1 \sim 1.3$ meV). We have experimentally verified that the shapes of signals do not change from small signals to large signals until close to the maximum saturation level.

In the low-temperature range ($0.2 \lesssim T/T_c \lesssim 0.3$), samples were immersed in superfluid. There are three phonon transmissivities[7] to be determined, i.e., $\eta_{\rm He}$ for the Pb—superfluid-He interface, $\eta_{\rm Q}$ for the Pb-quartz substrate interface, and $\eta_{\rm ox}$ for the Pb-PbO$_x$ interface.

As discussed in the previous section, in the low-temperature range, the χ's and γ's in Eqs. (16) and (17) can be related to the η's by the following equations:

$$\chi_1 = \eta_{\rm ox} v_s / 8 d_1 \ ,$$

$$\chi_2 = \eta_{\rm ox} v_s / 8 d_2 \ ,$$

$$\gamma_1 = \eta_{\rm He} v_s / 8 d_1 \ ,$$

$$\gamma_2 = \eta_{\rm Q} v_s / 8 d_2 \ ,$$

where d_1 and d_2 are the film thicknesses for films 1 and 2, respectively.

For sample 6 listed in Table I, $d_1 = d_2 = 500$ Å. We have experimentally determined that the signal decays exponentially with a time constant of about 2.5 nsec for either film 1 or film 2 irradiated by laser pulses. We cannot completely determine the three η's based on the information obtained for this sample alone because we have only two equations to solve for the three parameters. But we can draw certain conclusions immediately. Because the signal is only proportional to the sum of the excess quasiparticle densities and $d_1 = d_2$ for this sample, the signal does not change at all if the quasiparticles are transferred from one film to the other by the 2Δ phonon transmission through the Pb oxide. Therefore the fact that the signal decay times are approximately the same for either film 1 or film 2 irradiated by light implies that either $\eta_{\rm Q} \approx \eta_{\rm He}$ or $\eta_{\rm ox}$ is much larger than both $\eta_{\rm Q}$ and $\eta_{\rm He}$. Which of these two cases is true can be determined by the result of sample 7. For sample 7 with $d_1 = 500$ Å and $d_2 = 2500$ Å, we have observed very different signal shapes. The signal decays much slower when film 2 (2500 Å) is irradiated by light and the signal decays at least equally fast as the signals for sample 6 when film 1 (500 Å) is irradiated. This fact tells us immediately that $\eta_{\rm ox}$ cannot be much larger than either $\eta_{\rm Q}$ or $\eta_{\rm He}$, otherwise the two decay times for sample 7 should also be approximately the same. With this additional information, we can determine the decay time constant for sample 6 from the Eqs. (18) and (21). It is easy to show that for $\gamma_1 = \gamma_2 \equiv \gamma$ and $\chi_1 = \chi_2 \equiv \chi_3$, we have

$$R_{\pm} = \begin{cases} 2\chi + \gamma \\ \gamma \end{cases} \ ,$$

and $A = 0$, $B = 1$, so that the signal decays exponentially with a single decay time γ^{-1}. Since

$$\gamma^{-1} = 2\tau_\gamma = 2(4d/\eta v_s) \ ,$$

we conclude that $\eta_{\rm Q} \approx \eta_{\rm He} \approx 0.16$ for v_s chosen to be 10^5 cm/sec which is appropriate for the transverse phonons in Pb.

In the high-temperature range ($0.7 \lesssim T/T_c \lesssim 0.9$), $\gamma_1 = 0$ because the samples were in vacuum. Since there is no reason for the phonon transmissivity to change drastically from the low-temperature to high-temperature range, we use the same $\eta_{\rm Q}$ which has been determined by the low-temperature data. Then we have

$$\chi_1 = \eta_{\rm ox} v_s / 4 d_1 \ ,$$

$$\chi_2 = \eta_{\rm ox} v_s / 4 d_2 \ ,$$

$$\gamma_1 = 0 \ ,$$

$$\gamma_2 = \eta_{\rm Q} v_s / 4 d_2 \ .$$

for sample 1 ($d_1 = d_2 = 500$ Å) in Table I, we have experimentally observed a 3.1-nsec decay time for the signals with film 1 irradiated by laser pulses. It is easy to verify from Eqs. (18) to (21) that we have

$$R_{\pm}=\frac{1}{2}\{(2\chi+\gamma)\pm[(2\chi+\gamma)^2-4\gamma\chi]^{1/2}\}\ ,$$

$$A=-R_-/(R_+-R_-)\ ,$$

$$B=R_+/(R_+-R_-)\ ,$$

where $\chi\equiv\chi_1=\chi_2$ and $\gamma\equiv\gamma_2$.

Therefore the dominant time constant in Eq. (18) is R_- because $B>|A|$. γ^{-1} is equal to 1.25 nsec with η_Q being 0.16. Then χ can be determined by equating R_-^{-1} to 3.1 nsec. The result gives $\chi^{-1}\approx 1$ nsec which implies that $\eta_{ox}\approx 0.2$. The phonon transmissivities determined for the three interfaces are listed in Table II.

TABLE II. The phonon transmissivities for the Pb-quartz, Pb-superfluid He, and Pb-PbO_x interfaces and the corresponding phonon escape time for a 1000-Å Pb film.

	Pb-quartz	Pb-superfluid He	Pb-PbO_x
Phonon transmissivity η	0.16 ± 0.04	0.16 ± 0.04	0.2 ± 0.1
Phonon escape time for a 1000-Å Pb film (nsec)	2.5	2.5	2.0

In order to see whether this set of η's gives results consistent with the data for all samples listed in Table I, Figs. 3(a) and 3(b) show the comparison of the theoretical curve to the experimental traces. For more accurate comparison, we have convoluted Eq. (18) with a Gaussian response function with 1 nsec

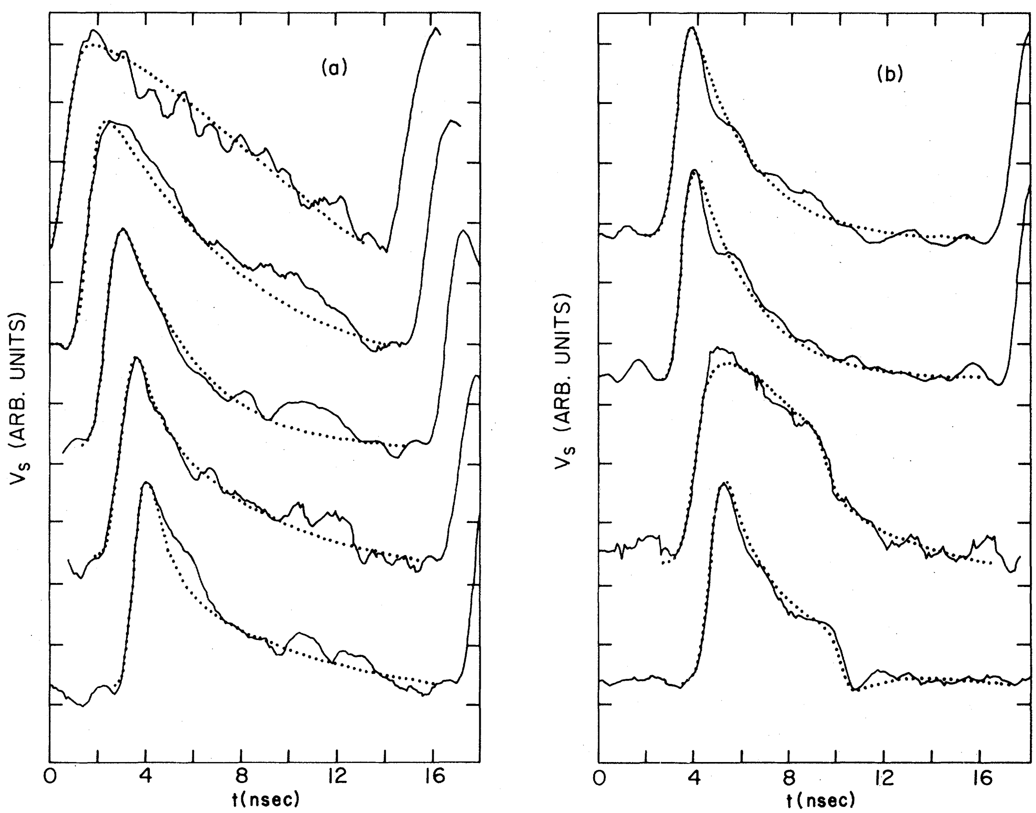

FIG. 3. (a) Voltage pulse V_s (solid lines) vs time t in high-temperature range ($0.7\lesssim T/T_c\lesssim 0.9$) for samples 3 (4000 Å, 4000 Å), 4 (1000 Å, 1000 Å), 1 (500 Å, 500 Å), 2 (500 Å, 1500 Å), and 5 (500 Å, 2500 Å), respectively, from the top to the bottom. Film 1 was irradiated by laser pulses for each case. There was no observable delay between the laser pulse and the voltage response V_s. For clarity, the time origins of the curves were displaced. (b) The voltage pulse V_s (solid lines) vs time t in the low-temperature range ($0.2\lesssim T/T_c\lesssim 0.3$) for samples 6 and 7. The first two (from the top to the bottom) correspond to the case of film 2 and film 1 of sample 6 irradiated by laser pulses, and the last two correspond to the case of film 2 and film 1 of sample 7 irradiated by laser pulses. The dotted lines in both (a) and (b) are theoretical fits with the phonon transmissivities for various interfaces given in Table II. A 25% negative reflection of the signal due to a loose connection of the coaxial cable to the amplifiers was observed for sample 7 [the last two curves in (b)]. The theoretical fitting curves for this sample have taken into account the 25% reflection.

half-width to simulate our experimental detection resolution. One can see that the set of η's listed in Table II gives a consistent fit to all the data. By changing the values of η's slightly, we conclude, from comparing the result with the experimental traces, that the variations cannot be more than ±20% in η_Q and η_{He}, and ±50% in η_{ox} and give a satisfactory fit to all the experimental traces. For the judgement of the curve fitting quality, we would like to make the following comments. Because of the limit of the available 14-nsec interval due to the high repetition rate of laser pulses, the data for thinner samples result in more stringent requirements on the curve fitting because the relaxation times are shorter. The curve fitting for the data taken in the high-temperature range is less stringent than for those in the low-temperature range. Unlike the situation for the sample immersed in superfluid He, for the sample in vacuum there is an additional dc voltage shift in the signal caused by the laser. Without knowing exactly where the baseline for the ac signal is, we have to adjust the baseline of the theoretical curve to make a best fit. We interpret this additional dc voltage shift as the substrate heating based on the following experimental observations. The amount of the dc voltage shift for the same light power is independent of the film thicknesses of the junction. Approximately the same amount of dc voltage shift can be induced by a cw laser of the same average power, which implies that the time constant for this heating effect is much longer than 14 nsec. This additional dc voltage shift is negligibly small when the sample and the substrate are immersed in superfluid He. Since we have already proven that the "cooling" of the Pb film through the superfluid interface is approximately equal to that through the substrate interface, the additional dc voltage shift cannot be attributed to the film heating alone.

IV. CONCLUSION

With 30-psec laser pulses, we have experimentally measured the threshold power to drive a 500-Å-thick superconducting Pb film normal. The relative threshold power as a function of temperature can be fitted with a theoretical calculation based upon the Rothwarf-Taylor equations and an estimate of the branching ratio of quasiparticles to phonons generated by photon excitations. The absolute absorbed threshold power intensity, or the absorbed energy density per pulse, cannot be estimated accurately due to the uncertainty in the estimate of the focused laser spot size. Using our best judgement, we claim that the absolute absorbed threshold power intensity for a 500-Å Pb film at low reduced temperatures ($T/T_c \lesssim 0.7$) is 3 to 30 kW/cm^2, or equivalently, 20 to 200 mJ/cm^3 per pulse. Based on the specific-heat data for superconducting Pb,[27] we estimate that the energy density needed to heat a Pb film from low reduced temperature to T_c is about 100 mJ/cm^3. Therefore we cannot rule out the possibility that the Pb film is driven normal by the simple heating effect. With 40-μsec laser pulses, Testardi[11] has measured the threshold power intensity and the energy density absorbed per pulse for a 275-Å Pb film on a sapphire substrate to be 3 W/cm^2 and 44 J/cm^3. The much lower threshold power intensity and much higher energy density per pulse of Testardi's results indicate that the phonon-trapping effect plays an important role if the laser pulse width is much longer than the phonon escape time. The temperature dependence of the threshold power for a 1000-Å Pb film irradiated with 1-μsec laser pulses has been measured by Golovashkin *et al.*[17] Their result indicates that the threshold power drops less sharply to zero near T_c than our result. It is not clear at this moment whether this discrepancy can also be caused by the difference in the laser pulse width.

Based on the analysis of the Rothwarf-Taylor equations, we have shown that, using the parameters appropriate for Pb, the quasiparticle relaxation time at temperatures close to T_c ($T/T_c \gtrsim 0.7$) is actually the phonon escape time τ_γ rather than the effective quasiparticle recombination time $\tau_{\text{eff}} \equiv \tau_R(1+\tau_\gamma/\tau_B)$. We have also shown that at low reduced temperatures ($T/T_c \lesssim 0.3$) the relaxation of a large amount of excess quasiparticles can be approximated by a simple exponential decay function with a time constant $2\tau_\gamma$. This approximation breaks down when the excess quasiparticle number drops down to a value such that the instantaneous quasiparticle recombination rate is comparable to τ_γ^{-1}. Measuring the relaxation times of quasiparticles with Pb tunnel junctions in vacuum at temperature close to T_c ($0.7 \lesssim T/T_c \lesssim 0.9$), and in superfluid at low reduced temperatures ($0.2 \lesssim T/T_c \lesssim 0.3$), we have determined the phonon transmissivities for various interfaces as listed in Table II. The calculation of Kaplan[28] indicates that $\eta_Q = 0.21$ for longitudinal phonons and 0.045 for the transverse phonons. Since the density of states for the transverse phonons is much higher than for the longitudinal phonons at energies near 2Δ, we should compare our result with the calculation for transverse phonons. Thus our value for η_Q is a factor of 3 to 4 higher than Kaplan's calculated value. Our value for η_{He} is within the range of values ($0.2 \sim 0.5$) generally accepted for the metal film-superfluid interface[29]; but it is at the low end. It has been generally assumed that phonons can transmit through the thin oxide barrier of a tunnel junction rather readily. Our result for η_{ox} indicates that this is not true at all. It is interesting to note that Kaplan *et al.*,[30] with a three-film double-Pb junction sample irradiated by cw light, obtained a η_{ox} 4 to 10 times less than our present value.

In summary, we conclude that the relaxation times of Pb films or Pb tunnel junctions are simply limited by the phonon escape times, which are in the order of few nanoseconds for a 1000-Å Pb film. Making a Pb tunnel junction with a thin film on top of a bulk Pb cannot speed up τ_γ because the thin oxide is also a significant barrier for phonons. Therefore, seeking the possibility of speeding up the quasiparticle relaxation to its intrinsic value, experiments designed to let quasiparticle diffusion carry energy away rather than phonons are in progress.

ACKNOWLEDGMENTS

We would like to thank M. L. W. Thewalt and D. J. Wolford, Jr., for letting us use their picosecond laser laboratory to do this experiment. Particular thanks go to M. Thewalt for technical assistance and stimulating discussions. We would also like to thank R. B. Laibowitz for his general interest and support; we also thank S. M. Faris, J. T. Yeh, and S. B. Kaplan for fruitful discussions. This research was supported in part by the Office of Naval Research.

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[21]Initially, the direct photon-excited quasiparticles step down in energy by creating more quasiparticles through the screened electron-electron interaction with an energy dependent rate $\sim 2\pi E^2/\hbar E_F$, which is in the order of 10^{15} sec^{-1} for $E = 1$ eV and the Fermi energy $E_F = 10$ eV. After the quasiparticle number increases by about a factor of 10 and the average quasiparticle energy drops to 0.1 eV, the phonon emission rate of quasiparticles becomes faster than the electron-electron interaction rate. The phonon emission rate of a high-energy quasiparticle can be approximately given by $2\pi \int_0^E \alpha^2 F(E)\, dE/\hbar$, where $\alpha^2 F(E)$ is the product of the phonon density of states and the square of the electron-phonon interaction matrix element. This phonon emission rate is independent of quasiparticle energy as long as E is larger than the Debye energy (about 9 meV for Pb). Therefore it takes approximately 10 to 20 phonon emission steps for quasiparticles to come down to several meV in energy. Each step takes approximately 0.025 psec according to the $\alpha^2 F(E)$ for Pb obtained by J. M. Rowell, W. L. McMillan, and R. C. Dynes (unpublished). Therefore we expect that 1 psec after the absorption of a photon, approximately 20 quasiparticles and 200 to 400 phonons are generated.

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