Quasiparticle and phonon lifetimes in superconducting Pb films

F. Jaworski and W. H. Parker
Department of Physics, University of California, Irvine, Irvine, California 92717

S. B. Kaplan
Department of Physics and Laboratory for Research on the Structure of Matter, University of Pennsylvania, Philadelphia, Pennsylvania 19174

(Received 3 March 1976)

The effective quasiparticle recombination lifetime in superconducting Pb films has been measured using Pb-oxide-Pb tunnel junctions and is consistent with the assumption that phonon lifetimes dominate the quasiparticle density relaxation. No evidence is found of the inelastic phonon processes suggested by Schuller and Gray.

In recent years there has been considerable experimental effort directed toward the determination of effective quasiparticle lifetimes in superconducting materials such as[1,2] Al and[3-6] Sn. While other materials have been investigated theoretically,[7] experimental data on quasiparticle lifetimes in other materials have been lacking until recently, except for a crude estimate of the lifetime in Pb.[8] Schuller and Gray[9] recently reported an experimental estimate of the effective quasiparticle lifetime in superconducting Pb obtained from measurements on optically illuminated Sn–Sn-oxide–Pb tunnel junctions. They concluded that while the phonon escape parameter from Pb to Sn agreed with calculated values, there was an indication of a more efficient and unexplained phonon-loss mechanism within the lead films. We report a measurement of the effective quasiparticle lifetime in superconducting Pb in agreement with a value calculated assuming the dominant phonon-loss mechanism is escape into the surrounding superfluid He. We find no evidence for the inelastic phonon processes suggested by Schuller and Gray. We also present a few comments on the analysis in their paper.

EXPERIMENT

The effective quasiparticle lifetime in Pb has been determined from measurements on optically illuminated Pb–Pb-oxide–Pb tunnel junctions using the indirect techniques developed earlier for measurements on Sn.[5] The rate equations describing the quasiparticle and phonon densities in a superconductor interacting with an external quasiparticle creation mechanism such as optical photons are

$$\frac{\partial N}{\partial t} = I_0 + \beta N_\omega - RN^2, \tag{1}$$

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

where we are using the same notation as Schuller and Gray. N is the number density of quasiparticles, I_0 is the volume rate of creation of quasiparticles by the external mechanism, N_ω is the number density of phonons with energy $\hbar\omega \geq 2\Delta$, β is the mean rate at which these phonons create quasiparticles, R is the mean rate of recombination, γ is the mean rate at which phonons of energy $\hbar\omega \geq 2\Delta$ are lost by processes other than quasiparticle creation, and $N_{\omega T}$ is the thermal number density of phonons. Terms in Eq. (2) corresponding to the processes of the emission and absorption of phonons by quasiparticles that do not result in a change of the quasiparticle numbers have been neglected.[10]

The steady-state solution of Eqs. (1) and (2) for the excess number density of quasiparticles is

$$n = \frac{N - N_T}{4N(0)\Delta_0} = \frac{N_T}{4N(0)\Delta_0}\left\{\left[1 + \frac{I_0\tau_R}{N_T}\left(1 + \frac{\beta}{2\gamma}\right)\right]^{1/2} - 1\right\}$$
$$= \frac{N_T}{4N(0)\Delta_0}\left[\left(1 + \frac{I_0\tau_{\text{eff}}}{N_T}\right)^{1/2} - 1\right], \tag{3}$$

where we have used the relation $\beta N_{\omega T} = RN_T^2$ obtained in thermal equilibrium with $I_0 = 0$. N_T is the thermal-equilibrium number density of quasiparticles, $N(0)$ is the single-spin density of states at the Fermi level, $\tau_R \equiv 1/RN_T$ is the mean quasiparticle recombination lifetime, and $\tau_{\text{eff}} = \tau_R(1 + \beta/2\gamma)$ is the effective quasiparticle recombination lifetime. For small creation rates $(I_0\tau_{\text{eff}}/N_T \ll 1)$ Eq. (3) becomes $N - N_T = \frac{1}{2}I_0\tau_{\text{eff}}$. Hence the excess quasiparticle density recombination time in the small-creation-rate limit is $\frac{1}{2}\tau_{\text{eff}}$.

In order to measure n and hence τ_{eff} it is necessary to relate n to an observable property of the superconductor such as the energy gap $\Delta(n)$. The dependence of Δ on the normalized excess number density n can be calculated from either the Owen-Scalapino model[11] or the modified heating model[12] with essentially the identical result that

$$\Delta(n)/\Delta(0) \simeq 1 - 2n \tag{4}$$

for $n \lesssim 0.1$ and $T/T_c \ll 1$.

As demonstrated earlier,[5] a combination of current injection of quasiparticles to drive the superconductor into a nonequilibrium state and photoexcitation to probe the nature of that state permits a determination of τ_{eff}. The quasiparticle creation rate when a tunnel junction is both current biased and optically illuminated is $I_0 = 2I_B/ev + I_c$, where I_B is the bias current, v the volume of the tunnel junction, and I_c the volume photoexcitation rate. The factor of 2 arises because a single tunneling electron creates two excitations of energy Δ. If $I_c \ll 2I_B/ev$ and I_c is modulated, the resulting modulation of the energy gap [from Eqs. (3) and (4)] is

$$\delta\Delta = \tau_{eff}\left(1 + \frac{2I_B\tau_{eff}}{evN_T}\right)^{-1/2} \frac{I_c}{4N(0)}. \tag{5}$$

At low temperatures where

$$\tau_{eff} = \tau_0 T^{-1/2} e^{\Delta/kT},$$

$$N_T = 4N(0)(\pi\Delta_0 k/2)^{1/2} T^{1/2} e^{-\Delta/kT},$$

and

$$\Delta(T) = \Delta_0,$$

we obtain

$$\delta\Delta \propto T^{-1/2} e^{\Delta/kT}(1 + A\,T^{-1} e^{2\Delta/kT})^{-1/2}, \tag{6}$$

where

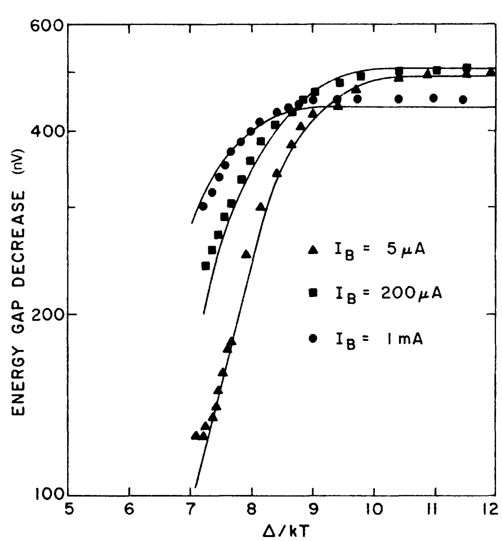

FIG. 1. Decrease of the energy gap vs Δ/kT for three different bias currents. The solid lines are Eq. (6) with the constant A adjusted for the best fit to the data.

TABLE I. Selected parameters of two of the curves in Fig. 1.

I_B (mA)	Total junction thickness (Å)	A (K)	τ_0 (sec K$^{1/2}$)
1.0	3542	1.2×10^{-6}	1.5×10^{-10}
0.20	3542	2.8×10^{-7}	1.8×10^{-10}

$$A = \frac{\tau_0 I_B}{2evN(0)(\frac{1}{2}\pi\Delta_0 k)^{1/2}} = \frac{\tau_0 I_B}{(3.03)v}. \tag{7}$$

The numerical value in Eq. (7) is for superconducting Pb, where we use $N(0) = 2.2\times10^{22}$ states/eV cm^3 and measure I_B in amperes and v in cm^3.

Figure 1 shows typical results for the measured temperature dependence of the modulation of the energy gap for Pb–Pb-oxide–Pb tunnel junctions at different bias currents. The junctions of area 0.34×0.35 mm^2 were fabricated on glass substrates, immersed in liquid helium, and biased by a high-impedance current source on the rapidly rising portion of the I-V curve at eV $=2\Delta$. The energy gap was optically modulated at 85 Hz by white light from a tungsten bulb and the resulting energy-gap modulation detected by a lock-in amplifier. The two curves in Fig. 1 corresponding to 200-μA and 1-mA bias current were obtained with the light intensity adjusted to satisfy $I_c \ll I_B/ev$. The solid curves are the best fit of Eq. (6) to the data using as adjustable parameters the constant A and a vertical scaling constant. The values of appropriate parameters and the calculated values of τ_0 are listed in Table I. Data from several other junctions yield values of τ_0 within ±30%.

The data in Fig. 1 obtained with a bias current of 5 μA illustrate the behavior when $I_c > I_B/ev$. Even though the data can still be fit by Eq. (6) as shown in the figure, a value of τ_0 cannot be extracted from the data. In order to obtain τ_0 using this analysis, the quasiparticle creation rate I_0 must be known. The creation rate from photoexcitation is not known.

All of the data are confined to the temperature region below 2.17 K. For reasons that are not understood at the moment, data obtained above 2.17 K cannot be explained with the analysis given here.

The average measured effective quasiparticle recombination time for several 3500-Å superconducting Pb tunnel junctions on glass substrates immersed in superfluid He is $\tau_{eff} = 2.06 \times 10^{-10} T^{-1/2} e^{\Delta/kT}$ ±30% and is in remarkable agreement with the calculated value of τ_{eff} assuming $\beta/2\gamma \gg 1$. In a superconducting thin film with a thickness d much greater than Λ, the phonon mean free path before pair breaking ($\Lambda \cong 400$ Å for a phonon of energy $\hbar\omega \cong 2\Delta$ in Pb), the phonon escape rate is $\gamma \cong \eta s/4d$. Here η is the transmissivity or probability of the phonon escaping from the film and s is the sound velocity. Assuming $\eta \cong 0.5$, as seems typical for superconductor-superfluid helium interfaces,[13] we calculate $\gamma \cong 3.9 \times 10^8$ sec^{-1}. Assuming $\beta/2\gamma \gg 1$ and using the value of β/R from Table II,

$$\tau_{\text{eff}} \cong \tau_R \beta/2\gamma = \beta/2R\gamma N_T = 1.9 \times 10^{-10} T^{-1/2} e^{\Delta/kT}.$$

The measured effective recombination lifetime is in remarkable agreement with the calculated value, but this must be considered accidental because of the uncertainties involved in calculating γ and β/R.

A theoretical value of τ_R calculated from the full $\alpha^2 F$ electron-phonon function obtained from electron tunneling and the method of Ref. 7 yields $\tau_R(\Delta_0) \simeq 2.0 \times 10^{-12} T^{-1/2} e^{\Delta/kT}$. When compared to the experimental value of τ_{eff} we obtain $\tau_{\text{eff}}/\tau_R \approx \beta/2\gamma \cong 100$, which easily satisfies the assumption $\beta/2\gamma \gg 1$.

DISCUSSION

Schuller and Gray's motivation for suggesting that strong inelastic phonon processes might represent the most efficient loss mechanism of high-energy phonons in Pb was the disagreement between their measured value of γ for Pb films and the earlier estimate of τ_{eff} in Pb by Parker and Williams.[8] The measurement of τ_{eff} reported here is approximately a factor of 25 larger than the earlier crude estimate and essentially removes any discrepancy in the experiments of Schuller and Gray. The definition of τ in Parker and Williams is smaller by a factor of 2 than the definition of τ_{eff} used here. The phonon lifetime for processes other than pair breaking, calculated assuming that escape into superfluid helium is the dominant phonon escape mechanism, is in essential agreement with both the results reported here and the results of Schuller and Gray. There is no longer any experimental need to suggest other phonon escape mechanisms.

The factor of 25 discrepancy between this and the earlier crude estimate of τ_{eff} in Pb is reduced to approximately 5 by the following considerations. First, Parker and Williams included only the volume of one of the superconducting films; both should be included because of the phonon coupling across the oxide barrier. Second, the bare density of states was used instead of the phonon-enhanced density of states $(1+\lambda)N(0)$. The source of the remaining factor of 5 discrepancy is unknown.

A detailed comparison between the results reported here and those of Schuller and Gray is of limited value because of a number of difficulties with their analysis. Since some of these difficulties are of general concern, we will comment in some detail on their analysis. In experiments on tunnel junctions of the kind under discussion here, only the sum of the energy-gap parameters can be measured. Hence Schuller and Gray used a curve-fitting parameter to fit expressions for $\Delta N = N - N_T$ of the illuminated film and $\Delta N'$ of the unilluminated film of their Sn–Sn-oxide–Pb tunnel junctions. These expressions were derived from rate equations similar to Eqs. (1) and (2) but including a term K_0 describing the number of phonons with energy $\hbar\omega \geq 2\Delta$ injected from external sources. A parameter ϵ defined by $\epsilon \equiv s'/s$, where $s \equiv (\Delta N)^2 + 2N_T \Delta N$, is introduced to describe the coupling between the two films. From the experiment they determine $\delta\Delta_{\text{expt}} = \delta\Delta + \delta\Delta'$ and relate this to ΔN by Eq. (4), obtaining $\delta\eta \equiv \Delta N + \Delta N' = 2N(0)\delta\Delta + 2N'(0)\delta\Delta'$. At this point Schuller and Gray make the approximation that the density of states at the Fermi level in Sn and Pb are equal. While it is true that the bare or band-structure densities of states of Sn and Pb are approximately equal, the number of quasiparticles depends on the phonon-enhanced density of states. The phonon-enhanced density of states can be calculated from the electronic specific heat[14] with the result that $N(0)$ of Pb is approximately 50% larger than that of Sn. It may be more appropriate to define $\epsilon' \equiv \epsilon[N(0)/N'(0)]^2$ and redefine

$$\delta\eta \equiv \frac{\Delta N}{2N(0)} + \frac{\Delta N'}{2N'(0)} \equiv -(\delta\Delta + \delta\Delta') = \delta\Delta_{\text{expt}}$$

before doing the analysis to obtain the coupling parameter ϵ. It is not obvious what change in ϵ would result.

Later in their analysis, Schuller and Gray calculate the number of phonons entering the unilluminated Sn film from the illuminated Pb film (K_0') but neglect any phonons entering the Pb film from the Sn (K_0). While, as shown below, this is a good approximation in their experiments, it may not be true in general. For example, the mean free path Λ in Sn of a longitudinal phonon of energy $\hbar\omega \cong 2\Delta_{\text{Pb}}$ may be equal to the film thickness of 2500 Å, in which case a sizable fraction of the phonons which leave the Pb film may return after passing through the Sn film and reflecting off the Sn-substrate interface.[15] The phonon transmissivity from Sn into Pb of approximately 1 ensures that most high-energy phonons returning to the Pb-Sn interface will pass into the Pb film.[16] One can conceive of other situations involving acoustically coupled superconductors where K_0 may not be negligible.

If the term K_0 is not neglected, Eq. (11) of Ref. 9 becomes

TABLE II. Some relevant physical parameters for Sn and Pb.

Parameter	Sn	Pb	Units
$N(0)$	1.4×10^{22}	2.2×10^{22}	$\mathrm{eV}^{-1}\,\mathrm{cm}^{-3}$
$\Delta(0)$	0.575×10^{-3}	1.365×10^{-3}	eV
$\overline{S}_T$ [a]	1.66×10^{5}	0.98×10^{5}	$\mathrm{cm\,sec}^{-1}$
$\overline{S}_L$ [a]	3.54×10^{5}	2.37×10^{5}	$\mathrm{cm\,sec}^{-1}$
$\overline{S}$ [b]	1.87×10^{5}	1.11×10^{5}	$\mathrm{cm\,sec}^{-1}$
$\beta/R = N_T^2/N_{\omega T}$	2.49×10^{19}	5.76×10^{18}	cm^{-3}
$\tau_{\mathrm{eff}}/(T^{-1/2}e^{\Delta/2kT})$	1.25×10^{-9}	1.4×10^{-10}	sec K
γ	9.3×10^{8}	5.5×10^{8}	sec^{-1}
Λ [c]	2.1×10^{3}	3.8×10^{2}	Å

[a] A simple average of $S_{\min}$ and $S_{\max}$ (Ref. 17).
[b] $3/\overline{S}^3 = 1/\overline{S}_L^3 + 2/\overline{S}_T^3$.
[c] For phonons with energy $\hbar\omega \cong 2\Delta$.

$$\epsilon = \frac{\beta' R}{\beta R'} \frac{\gamma_{\mathrm{PbSn}}}{\gamma'} \frac{\beta}{\gamma} \frac{1}{1 + \beta/2\gamma - 2K_0/(I_0 + 2K_0)} . \tag{8}$$

Since $2K_0/(I_0 + 2K_0) \le 1$ and $\beta/2\gamma \gg 1$ for Pb and Sn films immersed in liquid helium, Eq. (8) reduces to $\epsilon \cong 2(\beta' R/\beta R')(\gamma_{\mathrm{PbSn}}/\gamma')$ in the experiments of Schuller and Gray even if K_0 is not small. Thus the term K_0 could be of importance only in situations where $\beta/2\gamma \lesssim 1$.

A third difficulty arises in the calculation of the quantity $\beta/R = N_T^2/N_{\omega T}$. The calculation of N_T^2 contains the difficulty of obtaining the correct quasiparticle density of states, but of greater numerical consequence is the difficulty of obtaining the correct sound velocity required to calculate $N_{\omega T}$ from a Debye model. Within the Debye model $N_{\omega T} = (6k/\pi^2\hbar^3)(\Delta^2/s^3)Te^{-2\Delta/kT}$; thus $N_{\omega T}$ depends on the third power of the sound velocity.

Sound velocities in Sn span the range of a minimum transverse velocity of 1.3×10^3 m/sec to a maximum longitudinal velocity of 3.74×10^3 m/sec. In Pb the range of values is from 0.66×10^3 to 2.54×10^3 m/sec. We believe the correct value of the sound velocity to use in this calculation is the conventional average used with the Debye model, as shown in Table II. This value is also equal to that obtained from the Debye temperature. The resulting values for β/R shown in Table II differ from those used by Schuller and Gray by a factor of approximately 5 and are indicative of the obstacles preventing satisfactory comparison of one experiment with another (and with theory) on quasiparticle lifetimes in superconductors.

Our estimates of other significant parameters for Sn and Pb are shown in Table II. The values of γ and τ_{eff} are for 2500-Å-thick films immersed in liquid helium. The various experimental values of τ_{eff} for Sn have been scaled to 2500 Å and averaged to yield the value in the table; all experimental values are within a factor of 2 of this value. The phonon escape rate γ is calculated assuming a phonon transmissivity of 0.5 into liquid helium, neglecting any phonon escape from the other surface of the film, and assuming that the film thickness is larger than the phonon mean free path for pair breaking Λ. The estimate of Λ comes from the phonon lifetimes calculated in Ref. 7 and the average sound velocity of Table II. Because of uncertainties in the experimental data required for the calculations and because some of the quantities are energy dependent, such as Λ, it is unlikely that the values of β/R, τ_{eff}, Λ, and γ are more accurate than about a factor of 2. Such is the state of our understanding of quasiparticle and phonon lifetimes in superconductors.

[1] K. E. Gray, A. R. Long, and C. J. Adkins, Philos. Mag. 20, 273 (1969).
[2] L. N. Smith and J. M. Mochel, Phys. Rev. Lett. 35, 1597 (1975).
[3] W. Eisenmenger, *Tunneling Phenomena in Solids*, edited by E. Burstein and S. Lundquist (Plenum, New York, 1969).
[4] P. Hu, R. C. Dynes, and V. Narayanamurti, Phys. Rev. B 10, 2786 (1974).
[5] W. H. Parker, Solid State Commun. 15, 1003 (1974).
[6] G. A. Sai-Halasz, C. C. Chi, A. Denenstein, and D. N. Langenberg, Phys. Rev. Lett. 33, 215 (1974).
[7] S. B. Kaplan, C. C. Chi, D. N. Langenberg, J.-J. Chang, S. Jafarey, and D. J. Scalapino (unpublished).
[8] W. H. Parker and W. D. Williams, Phys. Rev. Lett. 29, 925 (1972).
[9] I. Schuller and K. E. Gray, Phys. Rev. B 12, 2629 (1975).
[10] J-J. Chang and D. J. Scalapino (private communication).
[11] C. S. Owen and D. J. Scalapino, Phys. Rev. Lett. 28, 1559 (1972).
[12] W. H. Parker, Phys. Rev. B 12, 3667 (1975).
[13] J. I. Gittleman and S. Bozowski, Phys. Rev. 128, 646 (1962).
[14] G. G. Gladstone, M. A. Jensen, and J. R. Schrieffer, in *Superconductivity*, edited by R. D. Park (Dekker, New York, 1969), Vol. II.
[15] C. C. Chi (private communication).
[16] The phonon transmissivity is obtained following the analysis of W. A. Little, Can. J. Phys. 32, 334 (1959).
[17] The minimum and maximum sound velocities of Pb are calculated from the elastic constants given in C. Kittel, *Introduction to Solid State Physics*, 4th ed. (Wiley, New York, 1971); the sound velocities of Sn are taken from J. A. Rayne and B. S. Chandrasekhar, Phys. Rev. 120, 1658 (1960).