Acoustic coupling of thin superconducting films*

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(Received 24 March 1975)

The acoustical coupling of two thin superconducting films forming a tunnel junction was measured. The experiment was performed by photoexciting quasiparticles in one film and studying the resulting change in the energy gaps. The results show that the phonon escape parameter from lead to tin agrees with calculated values and there is an indication of an additional escape mechanism in the lead films.

I. INTRODUCTION

In recent years there has been extensive work on nonequilibrium superconducting films to measure quasiparticle lifetimes[1-5] and to check theoretical calculations[6] about the nonequilibrium state.[4] The nonequilibrium state was obtained by increasing the number of quasiparticles above the thermal equilibrium value, and in most cases the role of the extra phonons was neglected. Although the importance of the phonons was recognized by Rothwarf and Taylor,[7] only a few attempts[8,9] were made to account for these because of the difficulty of measuring or adequately estimating their effect.

We have done an experiment similar to that of Parker and Williams[4] but which separates the effects of the phonons from that of the directly injected quasiparticles. Our results show that the phonon escape parameter from lead to tin agrees with calculated values, but more importantly, there appears to be another much more efficient escape mechanism in lead films which we shall discuss below.

We separate the contribution of directly injected quasiparticles from that due to an increase of the phonon population by a thin tunnel barrier between two optically opaque superconducting films. Phonons may readily pass this thin barrier (~20 Å thick), but the probability for electron transmission is very small. Experimentally we must find a way to obtain information about each film separately. The experiments performed by Parker and Williams[4] give us an insight as to how this can be done. They illuminate tin-tin and lead-lead junctions and find characteristically different temperature dependences, which are related to the different quasiparticle lifetimes in tin and lead. Through the use of a tin-lead junction, we can separate the contributions of the two films by their temperature dependence. This is not possible in a simple way using the same superconductors. In a junction of identical superconductors if each energy gap is perturbed differently, structure in the tunneling curves will appear at $\Delta_1 \pm \Delta_2$. Hence one could, in principle, separate the two contributions, but at low temperatures the weak structure at $\Delta_1 - \Delta_2$ is too hard to measure.

Our technique for determining the two contributions from the experimental data will be described in detail in Sec. II. We evaluate the temperature dependence for each film for various values of a coupling parameter ϵ and determine ϵ by choosing the best fit for the tin film, since its temperature dependence is much stronger than that of the lead. Finally, we interpret ϵ in terms of phonon escape parameters and discuss our conclusions.

II. THEORY

The equations that describe the nonequilibrium quasiparticle and phonon densities in a superconducting film[7] are

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

$$\frac{\partial N_\omega}{\partial t} = K_0 + \tfrac{1}{2}RN^2 - \tfrac{1}{2}\beta N_\omega - \gamma[N_\omega - N_\omega(T)] , \tag{2}$$

where N is the total number of quasiparticles, R is the recombination coefficient, N_ω is the total number of phonons with energy $\hbar\omega \geq 2\Delta$, $N_\omega(T)$ is the value in thermal equilibrium, β is the probability for pair breaking by such phonons, γ is the probability for phonons to be lost out of the energy range $\hbar\omega \geq 2\Delta$ by processes other than pair breaking, I_0 is the number of quasiparticles injected per cm^3 per sec, and K_0, a term introduced by us, is the number of phonons injected from external sources per cm^3 per sec.

Using the relation at thermal equilibrium, $\beta N_\omega(T) = RN(T)^2$, where $N(T)$ is the thermal-equilibrium quasiparticle density, the steady-state solution of Eqs. (1) and (2) is given by

$$S \equiv \Delta N^2 + 2N(T)\Delta N = (I_0/R)(1 + \beta/2\gamma) + K_0(\beta/\gamma R) , \tag{3}$$

where $\Delta N = N - N(T)$. Equation (3) is valid in both superconductors of a tunnel junction. However, if we illuminate one film of the junction, the nonilluminated film has no direct injection since the films are opaque, and then Eq. (3) becomes

$$S' \equiv \Delta N'^2 + 2N'(T)\Delta N' = K_0'(\beta'/\gamma' R') \ . \qquad (4)$$

In each equation the term on the right-hand side is the source perturbing the quasiparticle population. When it is zero, ΔN is zero.

The relative effect of the illumination on the nonilluminated film is determined by the ratio ϵ of the perturbing sources, i.e.,

$$\epsilon = \frac{S'}{S} = \frac{K_0' \beta'/\gamma' R'}{(I_0/R)(1+\beta/2\gamma) + K_0 \beta/\gamma R} \ , \qquad (5)$$

and this ϵ is the phonon coupling parameter that we determine experimentally.

Equations (3) and (4) are additionally coupled through the experimentally measured change in the energy gap ($\delta\Delta_{expt}$) due to illumination. In the theory of Owen and Scalapino,[6] for small values of $\delta\Delta/\Delta_0$ we have in each film $\delta\Delta = -\Delta N/2N(0)$, where $N(0)$ is the density of states at the Fermi energy in the normal metal, and Δ_0 is the energy gap at zero temperature. We measure $\delta\Delta_{expt} = \delta\Delta + \delta\Delta'$, and because the densities of states (both spins) of lead and tin are the same to within ~2% (1.81×10^{22} and 1.77×10^{22} $eV^{-1}\,cm^{-3}$, respectively), we use the very convenient simplification

$$\delta\eta \equiv \Delta N + \Delta N' \cong -3.6 \times 10^{22}\, \delta\Delta_{expt} \ , \qquad (6)$$

with $\delta\eta$ introduced for ease of notation. Using Eqs. (3)–(6) we obtain

$$\Delta N = \frac{1}{1-\epsilon}\left[\delta\eta + N'(T) + \epsilon N(T)\right] - \left[\left(\frac{\delta\eta + N'(T) + \epsilon N(T)}{1-\epsilon}\right)^2 - \frac{\delta\eta[\delta\eta + 2N'(T)]}{1-\epsilon}\right]^{1/2} \ , \qquad (7)$$

$$\Delta N' = \frac{-\epsilon}{1-\epsilon}\left(\delta\eta + N(T) + \frac{1}{\epsilon}N'(T)\right) + \left[\left(\frac{\epsilon[\delta\eta + N(T) + (1/\epsilon)N'(T)]}{1-\epsilon}\right)^2 + \frac{\epsilon\delta\eta[\delta\eta + 2N(T)]}{1-\epsilon}\right]^{1/2} \ , \qquad (8)$$

the other two solutions of the quadratic equations are not considered since they give nonphysical negative values for ΔN and $\Delta N'$. We can now evaluate these for given values of the parameter ϵ by using the BCS expression for $N(T)$ and the experimentally determined $\delta\eta$.

If we have sufficiently small injection of quasiparticles, we expect ΔN to depend on temperature[4] like $N(T)^{-1}$. In the lead film we never realize this in our experiment; however, for tin we do. We therefore determine the value of ϵ from the best fit of Eqs. (7) or (8) to $N(T)^{-1}$ for the tin film. These results will be presented and discussed later in this paper.

III. EXPERIMENT

The tunnel junctions were prepared under a pressure of 2×10^{-7} Torr with standard evaporation techniques. First, a 2500-Å tin film was condensed and a tunnel barrier formed by glow discharge oxidization[10] for 1 h under a pressure of 50 mtorr of oxygen. This was followed by depositing a 2500-Å lead film to form a tunnel junction of area ~0.04 mm^2 and having typical normal resistance of ≈1 Ω. For convenience we used glass instead of sapphire substrates, since Parker and Williams[4] have found no difference in similar optical excitation experiments at temperatures below the λ point of helium where our experiments were conducted. Immediately after evaporation the junction was mounted inside the cryostat and the cryostat was evacuated so as to expose the junction to air for as short a time as possible.

Light from a Spectra-Physics 15-mW He-Ne laser was mechanically chopped at 100 Hz and introduced in the cryostat through a $\frac{1}{8}$-in. fiber-optic bundle, illuminating one side of the tunnel junction. A small portion of the light was used as a lock-in signal for phase-sensitive detection. The intensity of the light was varied with a set of Oriel laser interference filters for the 6328-Å He-Ne line.

The junction was biased at the sum of the energy gaps at approximately the middle of the rising portion of the tunneling characteristic. The modulated voltage across the junction, which was equal to $\delta\Delta_{expt}$, was detected with a lock-in amplifier. The variation of the signal was recorded, while the bath temperature was allowed to rise slowly to the λ point after being first lowered to about 1.28 °K by pumping the helium. This process took about $\frac{1}{2}$ h, insuring a good approximation to thermal equilibrium. The smallest signals detected were of about 100 nV, whereas the average noise and pickup without illumination was less than 10 nV. Several checks at a chopping frequency of 500 Hz did not differ appreciably, and some runs were made several times to check consistency in the results. The experiment was repeated for several samples, with slightly different normal resistances, and the results obtained were essentially the same. This experiment was done also with a Ga-As injection laser, immersed in the liquid helium, and the results are in good quantitative agreement.

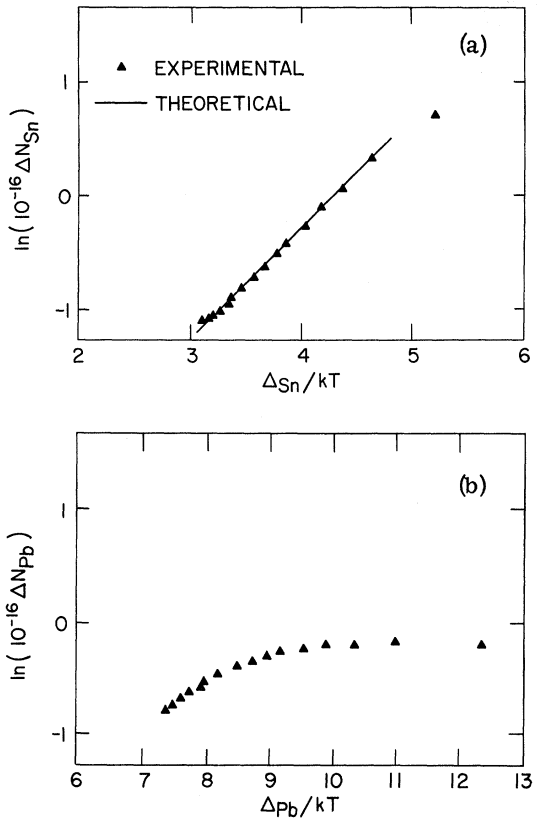


FIG. 1. Calculated values of (a) $\ln(\Delta N_{Sn})$ and (b) $\ln(\Delta N_{Pb})$ derived from the experiment with the tin illuminated at relatively low intensity. The value of ϵ is 0.04, and the solid line in (a) is a fit to the theoretical curve $N(T)^{-1}$ for tin.

IV. RESULTS AND CONCLUSIONS

The data were analyzed by using Eqs. (7) and (8) to evaluate ΔN and $\Delta N'$ for several values of ϵ from measured values of $\delta\Delta_{expt}$ as explained above. The slope of $\ln(\Delta N)$ or $\ln(\Delta N')$ vs Δ/kT or Δ'/kT was determined by fitting a straight line to these curves. It should be pointed out that the correct fit is not exactly a straight line, since $N(T)^{-1}$ is not an exact exponential but is multiplied by a factor $[T\Delta(T)]^{1/2}$. Over the temperature range of our experiment, this theoretical slope varies from 1.087 to 1.165; however, the scatter in the data made a closer fit of dubious use. An example of the curve ΔN_{Sn} vs Δ_{Sn}/kT for a particular value of ϵ is shown in Fig. 1(a), along with the theoretical curve $N(T)^{-1}$. Figure 1(b) shows an example of the curve ΔN_{Pb} vs Δ_{Pb}/kT for the same value of ϵ. It can be seen that the lead curve shows saturation. Figure 2 shows the value of the slopes for different ϵ's and for the various light intensities when lead is illuminated. For 0 attenuation the tin has a slope smaller than 1.0 for almost all values of ϵ because we are beginning to get saturation effects, with ΔN being of the order of $N(T)$. The mean value plus-minus one standard deviation from the mean, for ϵ is 1.1 ± 0.4. Figure 3 shows the values of the slope for different ϵ's for various light intensities when tin was illuminated and the mean value plus-minus one standard deviation from the mean, for ϵ is 0.05 ± 0.02. The nonzero value for ϵ, in this later case, indicates generation of phonons with energy larger than $2\Delta_{Pb}$, in the tin, so that pairs are broken in Pb. However, most of the phonons have energy $2\Delta_{Sn}$ and will not be able to break pairs in the lead [11,12]; so we will concentrate on the results with the lead illuminated.

We now wish to relate ϵ to phonon processes in the films. Assuming the phonons can escape the lead by various mechanisms, we write

$$\gamma = \gamma_{PbSn} + \gamma_{PbHe} + \gamma_{Pb}^{*} , \tag{9}$$

where γ_{PbSn} relates to phonons lost into the tin, γ_{PbHe} into helium, and γ_{Pb}^{*} to any inelastic processes in the lead film other than pair breaking. (Note that if the phonon energy is degraded below 2Δ it is effectively lost, [11,12] since it cannot break pairs.) The rate of phonons going from the lead film to tin is $\gamma_{PbSn}\,\Delta N_{\omega}$ [from Eq. (2)], and this is the only perturbation in the tin film, giving the term K_0' in Eq. (4). The steady-state solution of Eqs. (1) and (2) in the lead (if we neglect K_0) is

$$\gamma \Delta N_{\omega} = \tfrac{1}{2} I_0 . \tag{10}$$

So we write $K_0' = 2(\gamma_{PbSn}/\gamma)(I_0/2)$. The coupling of films via phonons is represented by K_0' and K_0, and since most of the phonons in the Sn are $2\Delta_{Sn}$ phonons, they cannot break paris in the Pb. Therefore we have neglected K_0 in arriving at Eq. (10). The extra factor of 2 accounts roughly for the fact

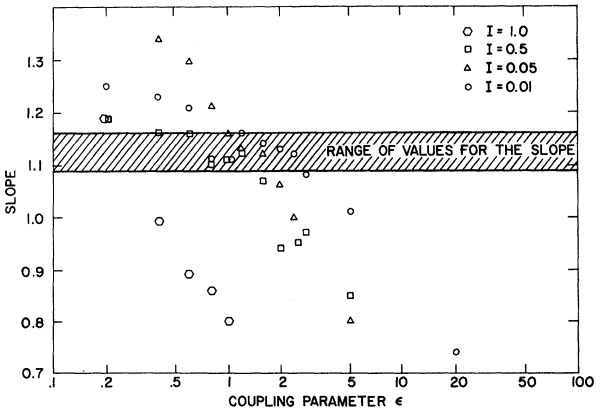
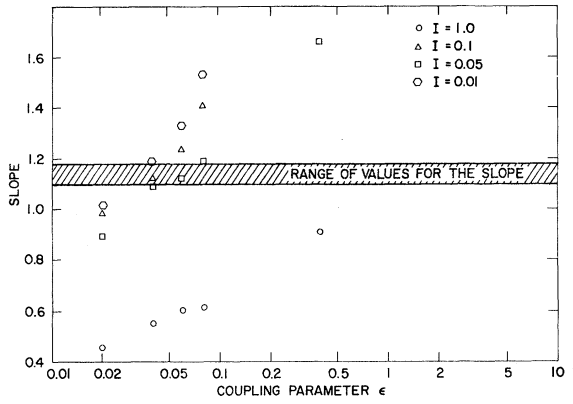


FIG. 2. Slopes of $\ln(\Delta N_{Sn})$ vs Δ_{Sn}/kT plotted as a function of the coupling parameter ϵ for the case when lead is illuminated for different relative illuminations. The dashed area indicates the theoretical values of the slope for low reduced temperatures.



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that $2\Delta_{Pb} > 2(2\Delta_{Sn})$, and two tin pairs may be broken by each $2\Delta_{Pb}$ phonon. Substituting this in Eq. (5), and neglecting K_0,

$$\epsilon = \frac{\beta' R}{\beta R'} \frac{\gamma_{PbSn}}{\gamma'} \frac{\beta}{\gamma} \frac{1}{1+\beta/2\gamma} \quad . \tag{11}$$

In order to simplify this expression, we examine the results of lifetime experiments in lead[4] and tin[5] which measure the recombination coefficient R_{exp}. This is related to R by

$$\frac{\beta}{1+\beta/2\gamma} = \frac{\beta}{R} R_{expt} \, . \tag{12}$$

Since β/R is known from statistics (we assume only transverse phonons to be important), these experiments are a measurement of $\beta/(1+\beta/2\gamma)$, and for $\beta/2\gamma \gg 1$ this is equal to 2γ, and for $\beta/2\gamma \ll 1$ this is equal to β. The measurements done in aluminum[3,8,9] and tin[5] indicate that $\beta/2\gamma \gtrsim 1$ and $\beta/2\gamma \gg 1$, respectively, and therefore in the following we expect $\beta/2\gamma \gtrsim 1$ and $\beta'/2\gamma' \gtrsim 1$. We solve the simpler case $\beta/2\gamma \gg 1$ and $\beta'/2\gamma' \gg 1$, since if $\beta/2\gamma \sim 1$ or $\beta'/2\gamma' \sim 1$, our results are different by factors of ~2, but the conclusions are still valid. With this, Eq. (11) becomes

$$\epsilon = 2 \frac{\beta' R}{\beta R'} \frac{\gamma_{PbSn}}{\gamma'} \quad ,$$

and since ϵ is measured and β/R and β'/R' can be determined from statistics, our experiment is a direct determination of γ_{PbSn}/γ'.

Table I summarizes the data we have used in this paper, and our measured lifetime in tin is within a factor of 2 with that measured by Ref. 5. Note that in the lead[4] and tin[5] lifetime experiments, the total thickness is about 3000 Å, corresponding closely to our lead and tin thicknesses of 2500 Å each. In our experiment when the lead is illuminated, the value of ϵ is 1.1 from which $\gamma_{PbSn}/\gamma' = 0.12$. Using γ' from Table I, we find $\gamma_{PbSn} = 7.7\times10^{8}$ sec^{-1}. But from the lifetime results on lead films,[4] $\gamma = 1.05\times10^{11}$ sec^{-1}, which is more than two orders of magnitude larger than our measured γ_{PbSn}. Although these lifetime experiments were done on sapphire substrates, one cannot expect a difference of 200 between phonon escape from lead into tin and into sapphire, nor would one expect γ_{PbHe} to be $200\,\gamma_{PbSn}$. This analysis suggests that the dominant escape mechanism is not to the substrate or helium but through the inelastic phonon processes mentioned in Eq. (9). We find in lead a phonon mean free path of 120 Å for these processes. According to the present understanding of anharmonicity, it does not seem reasonable even in a highly strained film to have such strong three-phonon processes. Inelastic collisions with defects expected to be roughly of this size cannot be ruled out.

We will now compare our experimental value of γ_{PbSn} with available calculations. We include the term γ_{Pb}^* and calculate the phonon mean free path inside the film as $\Lambda = 2s_T/(2\gamma_{Pb}^* + \beta)$, where s_T is the transverse sound velocity. If Λ is much less than the film thickness (i.e., escape to the edges is a small fraction of 2Δ phonons lost by all other processes), the calculation of the fraction of phonons escaping (f) in Ref. 8 gives γ_{PbSn}. If ΔS_ω is the rate of production of phonons per unit volume, then its source is the quasiparticles recombining given by $\Delta N/\tau = (I_0/2)(1+\beta/2\gamma)$, where τ is the true quasiparticle recombination time. The rate of phonons escaping is just the fraction escaping times the rate of production; so $\gamma_{PbSn}\Delta N_\omega = f\Delta S_\omega$. Using Eq. (10) together with the assumption that $\beta/2\gamma \gg 1$, we get $\gamma_{PbSn} = \frac{1}{2} f \beta$. In the lead

TABLE I. Relevant physical properties of tin and lead.

Parameter	Tin	Lead	Units
$N(0)$ (Ref. 1)	1.77×10^{22}	1.83×10^{22}	eV^{-1} cm^{-3}
Δ_0	0.60×10^{-3}	1.35×10^{-3}	eV
s_T (Ref. 14)	1.90×10^{5}	1.27×10^{5}	cm sec^{-1}
β_T/R	1.25×10^{20}	2.66×10^{19}	cm^{-3}
R_{expt} (Refs. 4 and 5)	1.04×10^{-10}	7.90×10^{-9}	cm^3 sec^{-1}
	$\beta/2\gamma \ll 1$		
β	1.30×10^{10}	2.10×10^{11}	sec^{-1}
Λ (pair breaking)	1460	60	Å
	$\beta/2\gamma \gg 1$		
γ	0.65×10^{10}	1.05×10^{11}	sec^{-1}

film (of thickness d), $d/\Lambda \gg 1$ and only the first term in the expansion of f is important so that $f = \alpha\Lambda/4d$, where α is the average phonon-transmission coefficient at the boundary. With the definition of Λ above,

$$\gamma_{\mathrm{Pb\,Sn}} = \frac{s_T}{4d} \frac{\alpha}{1+2\gamma_{\mathrm{Pb}^*}/\beta} \quad . \tag{13}$$

Since $\gamma \approx \gamma_{\mathrm{Pb}^*}$ and $\beta/2\gamma \gg 1$, we have $2\gamma_{\mathrm{Pb}^*}/\beta \ll 1$ so that $\gamma_{\mathrm{Pb\,Sn}} = 1.27\times10^9\alpha\ \mathrm{sec}^{-1}$. Comparing this with our measured value of $7.7\times10^8\ \mathrm{sec}^{-1}$, we find α at the lead-tin interface to be 0.61. Calculations by Little[13] give a value of about 0.15, and this is reasonably good agreement in view of the uncertainties about the microscopic nature of the interface and small numerical uncertainties when using the results of different experiments.

Finally the following additional comments support our conclusions. Preferential recombination of quasiparticles at the surfaces of the films would lead to a larger calculated λ, but our small value of measured $\gamma_{\mathrm{Pb\,Sn}}$ contradicts this possibility. Reconsidering the case $\beta/2\gamma \ll 1$, this is incompatible with the lifetime results[4,5] if phonons only escape the edges of the films. From Table I the transverse-phonon mean free path for pair breaking is 1460 Å for tin and 60 Å for lead. Using Fig. 2 of Ref. 8, these imply $\beta/2\gamma = 5.7$ in tin and $\beta/2\gamma = 63$ in lead, even with perfect acoustical coupling, which contradicts the original assumption of $\beta/2\gamma \ll 1$. In other words, if $\beta/2\gamma \ll 1$, then almost all the phonons escape the edges. However, with the pair-breaking mean free path less than the film thickness (2500 Å), this is impossible. Therefore, if $\beta/2\gamma \ll 1$ in either lifetime experiment, there must be a very strong loss of 2Δ phonons by inelastic scattering in the film, i.e., terms like γ_{Pb^*} in Eq. (9). In the latter case if $\beta/2\gamma \ll 1$ and $\beta'/2\gamma' \ll 1$, Eq. (11) becomes $\gamma_{\mathrm{Pb\,Sn}} = (\gamma/\beta)\gamma' \times 0.15$, and in this case the lifetime experiments [see Eq. (12)] determine β and β' rather than γ and γ'. Therefore for tin,[5] $\beta' = 1.30\times10^{10}\ \mathrm{sec}^{-1}$, and we would have $\gamma_{\mathrm{Pb\,Sn}} = (\gamma/\beta)(\gamma'/\beta')\times1.9\times10^9$ from our measurements. The calculated value of $\gamma_{\mathrm{Pb\,Sn}}$ is now given by Eq. (13) as

$$\gamma_{\mathrm{Pb\,Sn}} = 1.27\times10^9\alpha \frac{\beta}{2\gamma_{\mathrm{Pb}^*}} \quad ,$$

with $\alpha \leq 1$. Hence we would have poor agreement when both $\beta/2\gamma$ and $\beta'/2\gamma'$ are much smaller than one, and therefore this possibility was ruled out.

ACKNOWLEDGMENTS

The authors thank Donell Sain for the work done in the initial stages of this experiment. We thank R. P. Huebener for helpful discussions and for bringing the original work about laser irradiation to their attention. We would like to thank John Ketterson for stimulating discussions, and the assistance of R. T. Kampwirth is gratefully acknowledged.

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*Work performed under the auspices of the U. S. Atomic Energy Commission.

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