

Intrinsic and Experimental Quasiparticle Recombination Times in Superconducting Films

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Received 13 September 1976/Accepted 30 September 1976

Abstract. Experimental quasiparticle recombination lifetime data for superconducting Al, Sn, and Pb films are compared with calculations based on a ray acoustic model taking account of the film thickness dependence of the reabsorption of recombination phonons. Information on the true or intrinsic quasiparticle recombination lifetime obtained from these and other data is discussed.

PACS Code: 74

The apparent recombination decay time τ_{eff} of quasiparticles in superconducting films depends strongly on the reabsorption of phonons emitted in recombination. This influence has been treated in terms of a ray acoustical model [1] for phonon absorption and transmission to the film substrate. In the present article we compare this model with available experimental data. In Sec. 1 we discuss the results of the model calculation, in Sec. 2 possibilities of estimating the important quantity Λ_w , the phonon reabsorption mean free path. In Sec. 3 experimental τ_{eff} data for Al, Sn, and Pb are compared with calculation, together with a discussion of presently available τ_R data.

1. Results of the Phonon Trapping Model

The influence of phonon reabsorption can be discussed in terms of three physically significant ranges of the thickness dependence of τ_{eff} . For *Range 1* with $d \ll \Lambda_w$ (d : film thickness, Λ_w : phonon reabsorption mean free path) phonon-quasiparticle scattering as a loss process in the film volume can be neglected and the influence of reabsorption is expressed by

$$\frac{1}{\tau_{\text{eff}}} = \frac{1 - \cos \varphi_{\text{max}l}}{\tau_{Rl}} + \frac{1 - \cos \varphi_{\text{max}t}}{\tau_{Rt}} \quad (1)$$

In this limit phonon reabsorption is determined by the limiting angles $\varphi_{\text{max}l,t}$ of the total phonon reflection

at the film substrate boundary. The indices l and t refer to longitudinal and transverse phonons, respectively. τ_{Rl} and τ_{Rt} are the intrinsic quasiparticle decay time constants for longitudinal and transverse recombination phonon emission without reabsorption by pair-breaking. Equation (1) indicates that the film thickness d and the phonon transmission coefficients for angles $|\varphi| < \varphi_{\text{max}}$ have no influence on τ_{eff} . For *Range 2* with $d > \Lambda_w$ the calculation [1] results in a linear dependence of τ_{eff} on d

$$\frac{1}{\tau_{\text{eff}}} = \frac{1}{d} \cdot \frac{N_{\omega T}}{N_T} \cdot \left[\frac{\bar{T}_t}{c_l^2} + \frac{1}{2} \frac{\bar{T}_l}{c_t^2} \right] \left/ \left(\frac{1}{c_l^3} + \frac{1}{2c_t^3} \right) \right. \quad (2)$$

$N_T = 2N(0)[2\pi\Delta(T)kT]^{1/2} \exp[-\Delta(T)/kT]$ density of thermally excited quasiparticles with $N(0)$ being the density of Bloch states at E_F .

$$\begin{aligned} N_{\omega T} &= N_{\omega l T} + N_{\omega t T} \\ &= \left(\frac{1}{c_l^3} + \frac{2}{c_t^3} \right) \cdot \frac{2\Delta^2 kT}{\pi^2 \hbar^3} \cdot \left(1 + 2 \frac{kT}{2\Delta} \right) \cdot \exp\left(\frac{-2\Delta}{kT} \right) \end{aligned}$$

density of thermally excited longitudinal and transverse phonons with energy exceeding 2Δ , where c_l denotes the longitudinal sound velocity and c_t the transverse sound velocity.

$$\bar{T}_{l,t} = \int_0^{\pi/2} d\varphi \sin 2\varphi T_{l,t}(\varphi)$$

are the average values of the angle dependent film substrate boundary transmission coefficients $T_{l,t}(\varphi)$ for longitudinal and transverse phonons, respectively.

In (2) again phonon losses within the film volume are not contained. An estimate [1] of the influence of the dominant loss processes by phonon-quasiparticle scattering shows this approximation to be correct for $T < 0.3T_c$ and $A_w < d < 10A_w$. With further increased film thickness $d \gg A_w$ and $T > 0.5T_c$ phonon decay processes within the film volume lead to *Range 3* with comparatively small 2Δ -phonon transmission across the film-substrate boundary and dominant volume decay of phonons. τ_{eff} in this range approaches an upper limiting value with no further film thickness dependence resulting in

$$\frac{1}{\tau_{\text{eff}}} = \frac{1}{\tau_{Rl}(1 + A_{vl}/A_{wl})} + \frac{1}{\tau_{Rt}(1 + A_{vt}/A_{wt})} \quad (3)$$

where $A_{vl,t}$ denotes the mean free path for longitudinal and transverse phonons by volume decay processes without by pairbreaking, and $A_{wl,t}$ the mean free path for longitudinal and transverse phonons by pairbreaking.

Equation (3) corresponds directly to the results of Rothwarf and Taylor [2].

Since information on A_w , the phonon reabsorption mean free path is of importance in distinguishing between Ranges 1, 2, and 3, we will first discuss the relation between A_w and τ_R resulting from the detailed quasiparticle-phonon balance and the possibility of estimating A_w and τ_R from ultrasonic absorption data.

2. Recombination Time τ_R , Phonon Reabsorption Mean Free Path A_w and Ultrasonic Absorption

The recombination time τ_R can be calculated from the mean free path A_w for phonon reabsorption, and vice versa, using the detailed balance relation [1, 2]. In order to check τ_R values, as derived or estimated from τ_{eff} measurements, or the amount of phonon trapping, experimental information on A_w —if not otherwise determined—appears possible by extrapolating ultrasonic absorption data. Taking account of both phonon modes the detailed balance relation [1, 2] together with

$$A_{wl,t} = c_{l,t}^4 \cdot \tau_{wl,t},$$

where $\tau_{wl,t}$ is the longitudinal or transverse phonon lifetime for reabsorption by Cooper-pair breaking,

results in

$$\frac{1}{\tau_R} = \frac{1}{\tau_{Rl}} + \frac{1}{\tau_{Rt}} = \frac{4}{N_T} \cdot \frac{N_{\omega T}}{\tau_w} \quad (4)$$

$$\tau_{Rl,t} = \frac{N_T}{c_{l,t}^4} \frac{A_{wl,t}}{N_{\omega l,t} T} = \frac{N_T \cdot \tau_{wl,t}}{N_{\omega l,t} T \cdot c_{l,t}^4}$$

with $\tau_{Rl,t}^{-1}$ being the recombination rate with longitudinal or transverse phonon emission,

$\tau_{wl,t}$ the longitudinal or transverse 2Δ -phonon lifetime, and

τ_w the average 2Δ -phonon lifetime.

For a linear frequency dependence of the electronic part of the ultrasonic absorption α , as valid [3] (for longitudinal phonons) in the limit $q \cdot l > 1$ (q : phonon wave number, l : electron mean free path), the reabsorption mean free phonon path can be expressed by

$$A_{wl,t} = \frac{4.34h}{1.57 \cdot K_{l,t} \cdot 2\Delta(0)} \quad (5)$$

with

$$K_{l,t} = \frac{\alpha_{l,t}}{\nu} [\text{dB cm}^{-1} \text{ s}] \quad (6)$$

and h being Planck's constant and ν the frequency. The factor 4.34 in the numerator of (5) relates the mean free phonon path to the intensity decay factor e^{-1} of the corresponding sound wave, while the denominator constant of 1.57 refers [4] to the ratio of $\alpha_s/\alpha_n = 1.57$ at $T/T_c < 0.5$ for the phonons of energy equal to 2Δ where

α_n is the normal conductor ultrasonic absorption constant, and

α_s the superconductor ultrasonic absorption constant. From (4), (5), (6) together with N_T , $N_{\omega l T}$, $N_{\omega t T}$ we obtain

$$\frac{1}{\tau_R} = \frac{18.37\Delta^3 \sqrt{\pi}}{h^4 N(0)} \sqrt{\frac{kT}{2\Delta}} \cdot \left(1 + 2 \frac{kT}{2\Delta} + \dots\right) \left(\frac{K_l}{c_l^2} + \frac{K_t}{c_t^2}\right) \exp\left(\frac{-\Delta}{kT}\right). \quad (7)$$

Equation (7) relates τ_R to the constants K_l and K_t which can be obtained from ultrasonic absorption data in the MHz frequency range according to (6). Comparing (7) with theoretical results [5–7] and taking account only of normal processes agreement is found with respect to the temperature dependence of τ_R and also with respect to the energy gap dependence $\tau_R \sim \Delta^{-3}$ for constant kT/Δ .

The linear frequency dependence of the electronic part of ultrasonic absorption both for longitudinal and transverse phonons expressed by (6) holds for normal processes in the range $q \cdot l > 1$ (see [8]). In ultrasonic measurements with high-purity single crystals and also in clean metal superconducting tunneling junctions the condition $q \cdot l > 1$ can easily be fulfilled. In determining the deformation-potential part of ultrasonic absorption the longitudinal wave contribution is directly obtained from the total electronic attenuation while in the evaluation of the transverse wave contribution electronic absorption in the MHz frequency range by electromagnetic interaction and phonon collision drag has to be subtracted from the total absorption [8]. These contributions are comparatively small in the high-frequency range of 2Δ -photons [3], in contrast to possible deformation-potential interaction via Umklapp-processes. Thus, an extrapolation of the ultrasonic α_t may become difficult. Umklapp-processes will also modify the extrapolation of α_t . Furthermore, it has to be noted that for K_l and K_v , averages of $\alpha_{l,t}$ over different crystal orientations must be used. Typical values of $K_{l,t}$ determined from ultrasonic absorption in Al, Sn, and Pb range from 0.2 to 0.5 [$\text{dB cm}^{-1}\text{MHz}^{-1}$]. With this in mind τ_R estimates from ultrasonic data by use of (7) provide upper limiting values.

3. Comparison of Experimental and Theoretical Data for τ_{eff} and τ_R

The discussion of τ_{eff} measurements in tunneling junctions or films reported by different authors requires information on the substrate material and crystal orientation as well as knowledge of whether the sample has been in direct contact with liquid ^3He or ^4He (see [9]). Also for measurements in vacuum, clean surface conditions are imperative [9, 10]. An additional experimental check of the reliability of the results is provided by the verification of the temperature dependence $\tau_{\text{eff}} \propto \sqrt{T} \exp(\Delta/kT)$ which is not found in the case of magnetic flux trapping or overinjection. Some authors prefer to report their data in terms of $\tau_R^* = 2\tau_R$ and $\tau_{\text{eff}}^* = 2\tau_{\text{eff}}$, i.e. "lifetimes" instead of the experimental "decay time" constants [2], as chosen in [1], where τ_{eff} has been used corresponding to the directly observable decay in pulse experiments. For comparison with literature data we also use now τ_R^* and τ_{eff}^* . For ease of comparison the results of calculation and experiment are listed in Tables 2 and 3.

3.1. Aluminum

The steady state measurements of Miller and Dayem [11], Levine and Hsie [12] using Al-I-Al-I-In sandwich structures on Al_2O_3 crystals substrates in direct contact with liquid ^3He are more difficult to compare with the phonon escape model than the experiments of Gray et al. [13], and Long [14] which have been performed with Al-I-Al junctions on Al_2O_3 crystals in vacuum. For the latter sample configuration we used the data listed in Tables 1 and 2 for calculating τ_{eff} in the thickness ranges $d \ll \Lambda_w$ and $d \geq \Lambda_w$ according to (1) and (2) without volume losses, i.e. $\Lambda_v \gg \Lambda_w$. For Range 1 with sample thickness $d \ll \Lambda$ we obtain from (1) for Al on Al_2O_3

$$\frac{1}{\tau_{\text{eff}}} = \frac{0.186}{\tau_{Rl}} + \frac{0.142}{\tau_{Rt}}$$

With $\tau_{Rl} > \tau_{Rt}$ as to be expected from theory [7, 15] we obtain $\tau_{\text{eff}} = 7 \cdot \tau_R$, i.e. phonon trapping cannot be less than by a factor of 7 for clean and plane Al junctions on Al_2O_3 in vacuum. Surface irregularities can reduce this trapping factor by diffuse phonon scattering [1].

For an estimate τ_R by the detailed balance relation we can use the value $\Lambda_{wt} = 3500 \text{ \AA}$, as obtained experimentally by Long [16] for the dominant transverse phonon contribution. Extrapolation of ultrasonic data of David [17] according to (5) and (6) results in average values of $K_l = 0.8$ and $K_t = 0.4$ [$\text{dB cm}^{-1}\text{MHz}^{-1}$] and $\Lambda_l = 3600 \text{ \AA}$, $\Lambda_t = 7200 \text{ \AA}$ for $\Delta = 200 \mu\text{eV}$ if the total normal state electronic absorption at MHz frequencies is taken into account. In view of the mentioned difficulties of this extrapolation the agreement between Long's data for α_t and the ultrasonic extrapolation appears satisfactory. In using $\Lambda_l = 3500 \text{ \AA}$ and $\Lambda_t = 7200 \text{ \AA}$ the τ_R calculation with (4) results in $\tau_{Rl} = 1.05 \times 10^{-7} \text{ s}$, $\tau_{Rt} = 2.7 \times 10^{-6} \text{ s}$ and finally in the intrinsic decay time $\tau_R = 1.04 \times 10^{-7} \text{ s}$ at $\Delta/kT = 4$ and $\Delta = 200 \mu\text{eV}$. The intrinsic recombination lifetime amounts to $\tau_R^* = 2.1 \times 10^{-7} \text{ s}$. Since $\tau_{Rl} \gg \tau_{Rt}$ is confirmed, the thin limit for τ_{eff} with a trapping factor of 7 results in $\tau_{\text{eff}} = 7.3 \times 10^{-7} \text{ s}$ or $\tau_{\text{eff}}^* = 1.45 \times 10^{-6} \text{ s}$.

So far the possibility of comparison with experimental data on thin film Al junction is limited. Long [14] reported lifetimes $\tau_{\text{eff}}^* = 1.4 \mu\text{s}$ and $1.5 \mu\text{s}$ measured by pulse decay with Al junctions 710 \AA and 950 \AA thick on Al_2O_3 , respectively, (with $\Delta = 200 \mu\text{eV}$ at $\Delta/kT = 4$) in agreement with our thin limit calculation. But also shorter lifetimes are observed with other similar samples. In these cases basically a crystalline surface

structure leading to diffuse phonon scattering could possibly account for a reduction of the limiting trapping factor [1]; also surface overlays of condensed residual gas have a similar effect [1]. In further measurements by Long [14] with samples of 1000 Å thickness with silicon monoxide overlay, lifetimes $\tau_{\text{eff}}^* = 0.45 \mu\text{s}$ were observed, indicating also enhanced phonon escape. Levine and Hsieh [12] reported decay times $\tau_{\text{eff}} = 0.2 \mu\text{s}$ at $\Delta = 4kT$ in Al junctions of 700 Å thickness with indium overlay in contact with liquid ^3He but without specifying the energy gap. In their sample configuration the In overlay and the ^3He contact destroys the limiting angle of total reflection in the Al junction. Consequently, values closer to our calculated intrinsic $\tau_R = 0.1 \mu\text{s}$ or $\tau_R^* = 0.21 \mu\text{s}$ are to be expected. The same argument applies to the results of Miller and Dayem [11], having obtained a decay time of 0.6 μs at $\Delta = 4kT$ with $\Delta = 220 \mu\text{eV}$, the Al junction thickness amounting to 500 Å.

In a recent steady-state experiment Smith and Mochel [18] determined the intrinsic lifetime τ_R^* for 900 Å Al films on glass. In this measurement the phonon trapping factor was directly obtained by alternating creation of quasiparticles via single particle tunneling and pairbreaking by phonons. From the different quasiparticle populations for the sample in vacuum or in contact with liquid ^4He the phonon trapping factor was evaluated with 8.6 ± 1.1 (vacuum condition). Since the acoustic properties of glass are not too far from Al_2O_3 this trapping factor shows reasonable agreement with the calculated thin limit value of 7 for Al on Al_2O_3 .

The intrinsic lifetime measured by Smith and Mochel [18] with $\tau_R^* = 1.4 \pm 0.1 \mu\text{s}$ at $\Delta/kT = 6$ reduces to $\tau_R^* = 0.154 \mu\text{s}$ at $\Delta/kT = 4$ in reasonable agreement with our estimated value of $\tau_R^* = 0.2 \mu\text{s}$.

Finally this number is to be compared with recent lifetime calculations of Kaplan et al. [19] using $\alpha^2(\Omega)F(\Omega)$ obtained from theory or tunneling measurements.

For Al and $\Delta/kT = 4$ their data result in $\tau_R^* = 0.88 \mu\text{s}$ about four times larger than our value. From this calculation [19] also the average phonon reabsorption mean free path results with the higher value of $A_w = 7500 \text{ Å}$ [20, 32].

Since most of the experimental data scatter within the limits of $\tau_R^* = 0.21 \mu\text{s}$ (following from $A_w = 3500 \text{ Å}$) and the calculated thin limit effective time $\tau_{\text{eff}}^* = 1.45 \mu\text{s}$ for Al junctions on Al_2O_3 ($\Delta = 4kT$ and $\Delta = 200 \mu\text{eV}$), still further careful experimental work also specifying surface structure and purity remains to be done in

this thickness region. From such measurements conclusive information on τ_R and A_w in Al is expected.

For the film thickness *Range 2* with $\alpha > A_w$ and proportionality between τ_{eff} and d some experimental data are available by the measurements of Long [14].

Our calculation using the data for Al on Al_2O_3 substrates in vacuum according to (2) with $A_v \gg A_w$ again for $\Delta = 4kT$ and $\Delta = 200 \mu\text{eV}$ results in: $\tau_{\text{eff}}^* = 0.108 \times d/[\text{cm}]$ [s]. Experimentally Long [14] obtained for several measurements in the thickness range $d = 1520 \text{ Å}$ to $d = 3540 \text{ Å}$ (reduced to $\Delta = 200 \mu\text{eV}$) $\tau_{\text{eff}}^* = 0.087 (\pm 9\%) \cdot d/[\text{cm}]$ [s]. The agreement [31] appears satisfactory since the average phonon boundary escape [21] values $\bar{T}_t = 0.20$ and $\bar{T}_l = 0.3$ used in the evaluation of (2) are also known only within 10% accuracy. This gives further support to Long's conclusion that his experimental results between $d = 1500 \text{ Å}$ and 3500 Å are well described within the linear thickness dependence regime of τ_{eff}^* on d . Apparently the acoustic boundary phonon escape model can be directly applied to evaporated pure Al films on Al_2O_3 and influences of grain boundaries or of the oxide barrier within the film on 2Δ -phonon scattering or propagation are not found. This is in agreement with Kinder's [22] result that the sound velocities in thin Al films as used in our evaluation agree with the velocities measured in bulk polycrystalline material. The linear thickness dependence of τ_{eff}^* in Long's measurements down to 1500 Å indicates the possibility of diffuse surface scattering [1], i.e. unchanged average value \bar{T} but increased total reflection angles φ_{max} ; or that in these films the mean free phonon path for transverse waves is smaller than 3500 Å. This is consistent with the transverse and longitudinal phonon resonances found by Kinder [22] in Al phonon detectors of film thickness 620 Å and 920 Å indicating a phonon mean free path in the range between 1000 Å and 3000 Å. The observed phonon interference in tunneling junctions of enhanced energy gap by oxygen background evaporation is in agreement with our suggestion that grain boundaries within the film and oxide tunneling barriers are not strong phonon scatterers. Also the results of Welte et al. [23] and Forkel et al. [24] on the high energy phonon escape from Al junctions are consistent with a phonon mean free path at $2\Delta_{\text{Al}}$ of about 2000 Å and no significant elastic phonon scattering within the film.

In order to resolve the question, whether the linear thickness dependence in the range 1500 Å to 3500 Å in Long's [14] measurements results from diffuse surface scattering of phonons or from A_w being less than

Table 1. Acoustic data used for the evaluation of (1) and (2)

	ρ [g · cm ⁻³]	$c_l \cdot 10^5$ [cm s ⁻¹]	$c_t \cdot 10^5$ [cm s ⁻¹]
Al	2.72	6.4 [42]	3.1 [42]
Sn	7.3	3.32 [42]	1.67 [42]
Pb	11.8	2.35 [43]	1.03 [43]
Al ₂ O ₃	4.0	11.0	6.04
c-axis			
Si	2.35	9.35	5.2
111			

3500 Å, further measurements with specified surface of the films are necessary.

Influences of bulk phonon absorption, i.e. independence of τ_{eff} on thickness at large d values in *Range 3* so far have not been found. Experimentally this would require the use of thin Al crystal plates or evaporated films of thickness exceeding $d=10000$ Å at temperatures $T > 0.5T_c$.

For completeness the calculated results for Al on Si with respect to *Range 1* and *2* are also introduced in Table 2. Corresponding experimental data are not available. The acoustical data of Si are listed in Table 1.

3.2. Tin

Measurements of τ_{eff} using pulse methods have been performed for Sn tunneling junctions on silicon substrates in vacuum and ⁴He, [9, 25] and also on Al₂O₃

substrates with ⁴He contact [26] and with additionally evaporated solid nitrogen layers [10]. τ_{eff} measurements using optical excitation with Sn on SiO₂ and Al₂O₃ in vacuum are reported by Sai-Halász et al. [27] and Parker [28].

For the small film thickness *Range 1* $d \ll \Lambda_w$ we obtain from (1) and the data in Table 1 and 2

$$\frac{1}{\tau_{\text{eff}}} = \frac{0.047}{\tau_{Rl}} + \frac{0.039}{\tau_{Rt}} \text{ for Sn on Al}_2\text{O}_3$$

and

$$\frac{1}{\tau_{\text{eff}}} = \frac{0.065}{\tau_{Rl}} + \frac{0.053}{\tau_{Rt}} \text{ for Sn on Si.}$$

With dominating transverse phonon contribution this results in thin limit trapping factors of 25.6 and 18.8 for Sn on Al₂O₃ and Sn on Si, respectively.

Phonon mean free path values for calculating τ_R range from direct $2A$ phonon reabsorption average values of $\Lambda_w \leq 700$ Å [29] to ultrasonic data [30] extrapolation with average values $\Lambda_l = 1600$ Å and $\Lambda_t = 2200$ Å or $\Lambda_t = 7300$ Å, accounting for the total transverse absorption or only for the deformation potential contribution, respectively, for the case of normal processes. From these different mean free path values it appears that ultrasonic data extrapolation for transverse phonons to frequencies at $2A_{\text{Sn}}$ does not describe the experimental limit $\Lambda_w \leq 700$ Å, this apparently results from dominant Umklapp contributions to phonon

Table 2. Numerical results of the phonon escape model and some experimental data for τ_{eff} in thickness Ranges 1 and 2 with various superconductor-substrate combinations

	Superconductor	Al		Sn		Pb		
		Substrate	Al ₂ O ₃	Si	Al ₂ O ₃	Si	Al ₂ O ₃	Si
Limiting angle of total phonon reflection in the film	$\varphi_{\text{max}l}$		35.6°	43.2°	17.6°	20.8°	12.3°	13.6°
	$\varphi_{\text{max}t}$		30.9°	36.6°	16.0°	18.7°	9.8°	10.9°
<i>Range 1</i> $d \ll \Lambda_w$ Eq. (1)	$1 - \cos \varphi_{\text{max}l}$		0.19	0.27	0.047	0.065	0.023	0.028
$\frac{1}{\tau_{\text{eff}}} = \sum_{l,t} \frac{1 - \cos \varphi_{\text{max}l,t}}{\tau_{Rl,t}}$	$1 - \cos \varphi_{\text{max}t}$		0.14	0.20	0.039	0.053	0.015	0.018
phonon trapping factor A $\tau_{\text{eff}} = A\tau_R$	A_{theor}		7	5	25.6	18.8	67	56
	A_{exp}		8.6 ^d	—	—	—	—	—
<i>Range 2</i> $d \geq \Lambda_w$ Eq. (2) average phonon boundary transmission factors	\bar{T}_l^c		0.3 ^a	0.46	0.084	0.12 ^b	0.043	0.063
	\bar{T}_t^c		0.2 ^a	0.34	0.068	0.094 ^b	0.026	0.037
linear thickness dependence B ; $\tau_{\text{eff}} = B \cdot d$	B_{theor} [s cm ⁻¹]		0.054	0.028	4.38×10^{-3}	3.48×10^{-3}	1.28×10^{-3}	0.88×10^{-3}
	B_{exp} [s cm ⁻¹]		0.044 ^e	—	4.2×10^{-3} ^f	3.30×10^{-3} ^g	—	—

References: ^a[21]; ^b[44]; ^c[45]; ^d[18]; ^e[14]; ^f[28, 10, 26] average, liq. He contact accounted for by a factor of 4; ^g[9, 25].

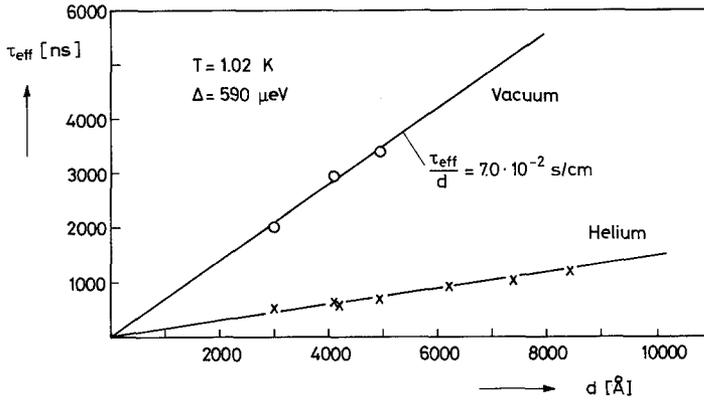


Fig. 1. Experimental recombination lifetime for Sn-junctions on Si-substrates [25] as function of film thickness. Pulse decay measurements in vacuum and also with liquid He contact. For $T=1.02$ K the vacuum measurement results in $\tau_{\text{eff}}/d=7.0 \times 10^{-2} \text{ s} \cdot \text{cm}^{-1}$. From this follows: $\tau_{\text{eff}}/d=3.3 \times 10^{-3} \text{ s} \cdot \text{cm}^{-1}$ at $T=\Delta/4k=1.71$ K (compare Subsec. 3.2). The experimental temperature dependence has been verified to follow the $\tau_{\text{eff}} \propto \exp(\Delta/kT)$ law

absorption. With $A_{wr}=700 \text{ \AA}$ and $A_{wl}=1600 \text{ \AA}$ the detailed balance equation (4) results in

$$\tau_{Rt}=6.75 \times 10^{-10} \text{ s} \quad \text{and} \quad \tau_{Rl}=1.08 \times 10^{-8} \text{ s}$$

for $\Delta=4kT$ and $\Delta=590 \text{ \mu eV}$. The transverse phonon contribution again dominating it follows that $\tau_R=6.36 \times 10^{-10} \text{ s}$ and $\tau_R^*=1.27 \times 10^{-9} \text{ s}$. Using the thin limit trapping factors this results in

$$\tau_{\text{eff, Sn/Al}_2\text{O}_3}=1.63 \times 10^{-8} \text{ s}$$

and

$$\tau_{\text{eff, Sn/Si}}=1.19 \times 10^{-8} \text{ s}$$

for Sn junctions with $d < A_w$ under vacuum conditions on Al_2O_3 and Si substrates, respectively.

Experimental results in this thickness range are available from the measurements of Sai-Halász et al. [27] for 400 \AA Sn films on quartz under vacuum conditions. The observed effective lifetimes reduced to $\Delta/kT=4$ are $\tau_{\text{eff}}^*=3.0 \times 10^{-8} \text{ s}$. From the limiting angle of total reflection for Sn on SiO_2 and the dominant transverse phonons we obtain according to (1) a trapping factor of 15. This results in $\tau_R^*=2.0 \times 10^{-9} \text{ s}$ from the measurement of Sai-Halász et al. reasonably close to our value $\tau_R^*=1.27 \times 10^{-9} \text{ s}$. In the theoretical work of Kaplan et al. [19] a somewhat longer lifetime of $\tau_R^*=4.6 \times 10^{-9} \text{ s}$ at $\Delta/kT=4$ was obtained together with a larger phonon reabsorption mean free path (dominant transverse phonons) of $A_w=1840 \text{ \AA}$ [20, 32]. Experiments with Sn tunneling junctions as phonon generators in contrast show strong reabsorption of relaxation phonons with energy exceeding 2Δ in accord with $A_w < 1000 \text{ \AA}$.

In most experiments on Sn [9, 10, 25–28, 33] the film thickness exceeded 1000 \AA and we expect a thickness dependence of τ_{eff} corresponding to Range 2 with $d > A_w$. Applying (2) with $A_w \gg A_w$ to the acoustical data of Sn on Al_2O_3 and Si results in

$$\tau_{\text{eff, Sn/Al}_2\text{O}_3}=4.38 \times 10^{-3} d/[\text{cm}] [\text{s}]$$

for Al_2O_3 substrates and

$$\tau_{\text{eff, Sn/Si}}=3.48 \times 10^{-3} d/[\text{cm}] [\text{s}]$$

for Si substrates at $\Delta=4kT$ and $\Delta=590 \text{ \mu eV}$. Experimental results by pulse decay measurements under vacuum conditions for Sn on Si [9, 25], compare Fig. 1, indicate the expected linear dependence of τ_{eff} on d .

From Fig. 1 the average slope of

$$\tau_{\text{eff}}=7.0 \times 10^{-2} d/[\text{cm}] [\text{s}]$$

for $T=1.02$ K and $\Delta=590 \text{ \mu eV}$ after reduction to $\Delta=4kT$ results in

$$\tau_{\text{eff, Sn/Si}}=3.30 \times 10^{-3} d/[\text{cm}] [\text{s}]$$

in good agreement with the calculated value $3.48 \times 10^{-3} d/[\text{cm}] [\text{s}]$. The total error between calculation and measurement amounts to $\pm 10\%$ corresponding to the limited accuracy of the calculation of \bar{T}_t and \bar{T}_i and the finite width of the energy gap.

We note that in this thickness regime the contribution of the longitudinal phonon escape amounts to 13% of the total phonon escape rate. The phonon trapping factor for the measurement with $d=3000 \text{ \AA}$ amounts to 140 comparing τ_{eff} with τ_R as obtained from $A_w=700 \text{ \AA}$. Since Fig. 1 shows no deviation from the linear thickness dependence down to $d=3000 \text{ \AA}$, the conclusion that A_w is smaller than 3000 \AA is consistent with $A_w=700 \text{ \AA}$. In contact with superfluid He the thickness proportionality of τ_{eff} in Fig. 1 is reduced by a factor of 4.6 indicating the strong deviation of the phonon escape [9, 34–36] into liquid ^4He from the acoustic model. For Sn on Al_2O_3 in contact with liquid He Parker [28] obtained the experimental value $\tau_{\text{eff}}=1.31 \times 10^{-3} d/[\text{cm}] [\text{s}]$ ($\Delta=4kT$) while pulse decay measurements [10, 26] with experimental data

$d=2300 \text{ \AA}$, $\tau_{\text{eff}}=8 \times 10^{-8} \text{ s}$ at $T=1.25 \text{ K}$ result in $\tau_{\text{eff}}=0.8 \times 10^{-3} d/[\text{cm}] [\text{s}]$ ($\Delta=4kT$). The difference of the experimental values exceeds the limits of $\pm 30\%$, as estimated in [28]. Comparing experiment with our calculated “vacuum value” $\tau_{\text{eff}}=4.38 \times 10^{-3} d/[\text{cm}] [\text{s}]$ ($\Delta=4kT$) the increase of the phonon escape rate by a factor of 3.3 to 5.5 agrees with the escape rate increase for Sn on Si in He. Finally, pulse decay measurements with optical excitation [33] of a Sn tunneling junction $d=3200 \text{ \AA}$ on glass in ^4He contact resulted in $\tau_{\text{eff}}=3.9 \times 10^{-8} \text{ s}$ ($\Delta=4kT$). This leads to

$$\tau_{\text{eff}}=1.22 \times 10^{-3} d/[\text{cm}] [\text{s}]$$

in agreement with the above data for Sn on Al_2O_3 in contact with liquid He. This is to be expected since the phonon radiation into the helium bath dominates.

An additional reduction of τ_{eff} in the linear thickness range has been observed by condensing nitrogen [10] on 2330 \AA Sn films on Al_2O_3 in contact with liquid He. The measured time constant is $\tau_{\text{eff}}=1.8 \times 10^{-7} \text{ s}$ at 1.02 K and corresponds to a trapping factor of about 13 as compared to τ_R with $A_w=700 \text{ \AA}$ at the same temperature. Evaluation by (2) indicates an increased phonon escape of about $\bar{T}_{t,l}=0.8$ or 80% by the condensed nitrogen-liquid He contact. Part of this influence is due to the limiting angles of total reflection in Sn increasing to 90° since the sound velocities in solid N_2 and liquid He are smaller than the sound velocities in Sn. Quantitatively the acoustic model, however, cannot account for the high phonon escape rate as is the case for the Kapitza resistance anomaly of liquid He. In contrast to this the acoustical model even at frequencies of 280 GHz corresponding to $2\Delta_{\text{Sn}}$ can be applied to the metal-solid boundary as is demonstrated by the close agreement between measurement and calculation for experiments under vacuum conditions.

Finally, also for the case of Sn no influence of volume absorption of phonons has been observed even at trapping factors exceeding 100, as indicated by the absence of saturation effects at larger d values in the $\tau_{\text{eff}}(d)$ dependence of Fig. 1. This result is of importance for the use of tunneling junctions as phonon generators [37] since the conclusion can be drawn that all recombination phonons leave the junction (under vacuum conditions) without volume losses. Further experiments with increased film thickness and temperature, i.e. in Range 3, are necessary to study the onset of volume losses and the possible application to phonon mean free path determination.

3.3. Lead

Although the first experimental and theoretical work [1, 5, 15] in order to determine τ_R has been concentrated on lead, only a few further measurements [39] have been performed. In the experiments samples in direct contact to liquid He were used. At 1.44 K the experimental value was $\tau_{\text{eff}}^* \leq 2.2 \times 10^{-7} \text{ s}$; the theoretical values are $\tau_R^*=4.3 \times 10^{-8} \text{ s}$ [5] and $\tau_R^*=1.67 \times 10^{-8} \text{ s}$ [15], both at 1.44 K , the latter also taking account of Umklapp processes. In [39] experiments on films with $1000\text{--}2000 \text{ \AA}$ thickness are reported which result in τ_{eff} values exceeding the theoretical result [15] by one order of magnitude.

For comparison of the experimental results with our model the acoustical data of Tables 1 and 2 are introduced in (1). For *Range 1* with $d < A_w$ we obtain

$$\frac{1}{\tau_{\text{eff}}} = \frac{0.023}{\tau_{Rl}} + \frac{0.015}{\tau_{Rt}} \text{ for Pb on } \text{Al}_2\text{O}_3 \text{ in vacuum}$$

and

$$\frac{1}{\tau_{\text{eff}}} = \frac{0.028}{\tau_{Rl}} + \frac{0.018}{\tau_{Rt}} \text{ for Pb on Si in vacuum.}$$

The trapping factors are 67 and 56 for Al_2O_3 and Si substrates, respectively, if only the dominant transverse-wave contribution is taken into account.

Ultrasonic total absorption [40] extrapolation results in values of $A_{wl}=2100 \text{ \AA}$ and $A_{wt}=770 \text{ \AA}$. From these follow by detailed balance (4) $\tau_{Rl}=2.1 \times 10^{-9} \text{ s}$ and $\tau_{Rt}=0.79 \times 10^{-10} \text{ s}$ for $\Delta=4kT$ and $\Delta=1.35 \text{ meV}$, i.e. $T=3.92 \text{ K}$, and the intrinsic decay time

$$\tau_R=0.72 \times 10^{-10} \text{ s}$$

or the lifetime $\tau_R^*=1.45 \times 10^{-10} \text{ s}$, result again with dominant transverse wave contribution. For comparison with experimental and theoretical data cited above we reduce our τ_R to τ_R^* at 1.44 K and obtain: $\tau_R^*=2.4 \times 10^{-7} \text{ s}$. The rough agreement with the experimental value [38] appears fortuitous, instead the theoretical values [5, 15], which are about one order of magnitude below our value, indicate again that the mean free path for transverse phonons is smaller than obtained from the ultrasonic extrapolation. This is in agreement with the phonon mean free path obtained from the data of Kaplan et al. [19] with $A_w=350 \text{ \AA}$. But disagreement exists with $\tau_R^*=3.9 \cdot 10^{-9} \text{ s}$ at $\Delta/kT=4$ as derived from their work. Since, assuming $A_w < 770 \text{ \AA}$, all experiments have been performed in *Range 2* with

Table 3. Intrinsic recombination lifetimes τ_R^* calculated from A_w estimates compared with experimental values and data of A and $N(O)$ used for the calculation

	Al	Sn	Pb
A [meV]	0.20	0.59	1.35
$N(O)$ [$\text{eV}^{-1} \text{cm}^{-3}$]	1.75×10^{22} ^a	1.42×10^{22} ^a	2.22×10^{22} ^a
A_{wt} [\AA] ($\Omega=2A$)	3500 ^b	700 ^c	770 ^d
A_{wt} [\AA] ($\Omega=2A$)	7200 ^e	1600 ^f	2100 ^d
τ_R^* [s] calc. from (4) with $A_{wt,t}$ ($A/kT=4$)	2.1×10^{-7}	1.27×10^{-9}	1.45×10^{-10}
τ_R^* [s] exp ($A/kT=4$)	1.54×10^{-7} ^g	2.0×10^{-9} ^h	—

References: ^a[46]; ^b[16]; ^c[29]; ^d[40]; ^e[17]; ^f[30]; ^g[18]; ^h[27] with a trapping factor of 15.

$d > A_w$, we now discuss $\tau_{\text{eff}}(d)$ according to (2). From the data of Tables 1 and 2 we obtain:

$$\tau_{\text{eff}} = 1.28 \times 10^{-3} d / [\text{cm}] \text{ [s]}$$

at $A/kT=4$ or $T=3.92$ K for Pb on Al_2O_3 and further $\tau_{\text{eff}}=0.88 \times 10^{-3} d / [\text{cm}]$ [s] for Pb on Si. For $T=1.44$ K and a film of 1250\AA thickness on Al_2O_3 , this results in $\tau_{\text{eff}}=5.9 \times 10^{-5}$ s, which exceeds the experiment [38] by two orders of magnitude.

We note that at 1 K τ_{eff} increases to $\tau_{\text{eff}}=9 \times 10^{-3}$ s as follows from the exponential temperature dependence. Since the exponential temperature law has not been checked in the measurements [38] we believe that strong overinjection [2] has lowered the experimental value as a consequence of a quasiparticle population highly increased above thermal equilibrium. This is substantiated by phonon pulse experiments [41] using lead junctions as phonon detectors which almost show no temperature dependence of τ_{eff} in the range of 1 K to 2 K. Also in optical excitation [39] the overinjection limit is easily reached. More measurements in lead are to be awaited in order to draw conclusions on the model and also on the question of volume which may be of special importance in this superconductor.

Conclusions and Summary

The influence of recombination phonon reabsorption and reemission on quasiparticle decay times can be described by an acoustical ray or ballistic phonon propagation model [1] with three characteristic ranges of the thickness dependence of the experimental quasiparticle density decay time $\tau_{\text{eff}}(d)$. In the limit of d small compared to the phonon reabsorption mean free path A_w only the limiting angles of total phonon reflection φ_{max} at the film boundaries determine the

phonon trapping factor or the ratio τ_{eff}/τ_R with τ_R the intrinsic or true quasiparticle decay time without phonon trapping. Experimental data for Al on Al_2O_3 substrates [14, 11, 12] with different outer surface conditions in this film thickness range scatter between $\tau_{\text{eff}}=\tau_R$ and $\tau_{\text{eff}}=7 \cdot \tau_R$. The trapping factor of 7 is calculated from the limiting angle of total reflection at the Al- Al_2O_3 boundary without contact to liquid He. Further results are listed in Table 2. For the film thickness range $d > A_w$ the model results in a linear increase of $\tau_{\text{eff}}(d)$ in the absence of volume losses, i.e. $A_d \gg A_w$. Experimental data for Al on Al_2O_3 [14] and Sn on Si [9, 25] agree with the constant of proportionality calculated from the model by use of the phonon and electron densities of state and the phonon escape rate across the film-substrate boundary, as obtained from acoustical data (see Table II). In addition, the quantitative determination of enhanced phonon escape by contact with liquid ^4He agrees with studies of the Kapitza resistance anomaly.

So far experiments in the linear $\tau_{\text{eff}}(d)$ range up to d -values of 5000\AA for Sn films with trapping factors of 240 have not indicated any influence of volume losses of phonons leading to a saturation of τ_{eff} at large film thicknesses $d \gg A_w$. From this follows that in phonon pulse experiments using superconductor tunneling junctions as generators under vacuum conditions all primary recombination phonons are finally radiated into the substrate crystal without losses, i.e. no “thermalisation” of recombination phonons takes place even at film thickness values far exceeding A_w . Experiments with extremely thick films are of interest with respect to the determination of volume losses and the possibility of τ_R calculations [1].

For the $\tau_{\text{eff}}(d)$ dependence of Pb films and other film substrate combinations, calculated results are presented in Table 2.

For Al, Sn, and Pb the intrinsic recombination time τ_R is evaluated from the phonon mean path A_w as obtained by the extrapolation of ultrasonic absorption or by direct estimates (see Table 3).

Acknowledgements. We gratefully acknowledge discussions with C. J. Atkins and A. R. Long, and are especially indebted to A. R. Long for privately communicating further experimental data. Financial support granted by the "Deutsche Forschungsgemeinschaft" to part of the experimental work is gratefully acknowledged.

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Note added in proof. The lifetimes in [18] have to be multiplied by a factor of 2.84. (Private communication by L. N. Smith)