

Quasiparticle Recombination Time in Superconducting Tin and Normal Electronic Density of States at the Fermi Surface from Tunnel Junction Experiments

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Received July 3, 1978

We have measured the temperature dependence of the effective quasiparticle recombination time in superconducting tin tunnel junctions by current and laser pulse excitation. The experimental times show satisfactory agreement with calculations based on the ray acoustic lifetime model of Eisenmenger et al. taking into account the film thickness dependence of the phonon reabsorption, 2Δ -phonon volume loss processes and phonon transmission from the junction into the substrate and liquid helium. On the basis of the BCS density of thermally excited quasiparticles and simplified rate equations for quasiparticle recombination, and from the analysis of measurements of decaying excess quasiparticle concentrations we obtain a mean value $N_0 = (2.73 \pm 0.03) 10^{22} \text{ eV}^{-1} \text{ cm}^{-3}$ for the electronic density of states at the Fermi Surface in thin, evaporated tin films. This value differs less than 5% from that obtained from the experimental electronic heat-capacity coefficient of the bulk material.

I. Introduction

The minimum energy difference between the ground state of a superconductor at a temperature below the transition temperature T_c and the lowest quasiparticle states corresponds with the BCS energy gap $\Delta(T)$. Thermally excited quasiparticles with energies $E \geq \Delta$ decay to the ground state within a characteristic time τ_R via the formation of Cooper-pairs and the emission of phonons of the energy $\hbar\omega \geq 2\Delta$. Whereas τ_R , the intrinsic recombination time, is not directly accessible by experiment, an effective time constant τ_{exp} can be determined from the decay of an excess quasiparticle concentration generated by an external dynamic creation mechanism, such as pair breaking by the absorption of phonons [1, 2] or photons [3–6] or by tunneling between superconductors [7–15]. As has been shown in several experimental [1–5, 10, 12–15] and theoretical [10, 16, 17] investigations, τ_{exp} significantly exceeds τ_R by the trapping of recombination phonons via repeated reabsorption and

emission of 2Δ -phonons within the superconducting device. The effectiveness of phonon trapping essentially depends on the sample thickness and the acoustic surface boundary conditions for phonon escape to the adjoining media, i.e. substrate material and liquid helium or vacuum, respectively [2, 15, 17].

Up to now two different experimental techniques have been applied for the determination of τ_{exp} , the indirect steady-state [3, 5, 7–10, 13–15] and the direct decay time [1, 2, 4, 6, 10–12] measurement. The steady-state method usually measuring τ_{exp} in a sandwich-structure of two tunnel junctions requires the knowledge of the density N_0 of electronic states at the Fermi level as resulting from heat-capacity measurements of the clean bulk material, see e.g. [8]; alternatively, also the band-structure quantity has been used [10, 14]. The samples for the τ -experiments, in contrast, consist of vacuum-deposited films of microcrystalline structure, usually with high disorder. Consequently, the question arises for the value of N_0 in disordered metal films.

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We report here on direct measurements of the effective quasiparticle recombination time τ_{exp} in superconducting tin and a new experimental technique to determine N_0 in thin superconductor layers. Using electron tunneling between superconductors for generating and probing an excess quasiparticle density under pulsed conditions, this method has also the advantage of being able to check directly the dependence of τ_{exp} on "over-injection". Moreover, we have investigated to what degree τ_{exp} differs from quasiparticle excitation by current pulse (excitation energy $E \simeq \Delta$) and laserpulse ($E \gg \Delta$) for the same sample. The measured temperature dependence of τ_{exp} is compared with that calculated from a geometrical-acoustic 2Δ -phonon trapping lifetime model [17]. Possible disturbing influences on the exponential shape of $\tau_{\text{exp}}(T)$ at low temperatures are studied.

II. Theory

The method to be described is based on the fundamental relationship

$$\delta N = N - N_{\text{th}} = I_0 \tau_{\text{exp}}, \quad (1)$$

between the excess quasiparticle density δN , the stationary injection or generation rate I_0 and τ_{exp} . According to Rothwarf and Taylor [16] τ_{exp} can be expressed by

$$\tau_{\text{exp}} = \left[\left(\frac{N_{\text{th}}}{I_0} \right)^2 + \frac{2N_{\text{th}}}{I_0} \tau_{\text{exp},0} \right]^{1/2} - \frac{N_{\text{th}}}{I_0}, \quad (2)$$

where $\tau_{\text{exp},0}$ is the experimental recombination lifetime related to $\delta N/N_{\text{th}} \ll 1$ and N_{th} the equilibrium concentration of thermally excited quasiparticles. The expression

$$\tau_{\text{exp}} = \tau_{\text{exp},0} / (1 + \delta N / 2N_{\text{th}}) \quad (3)$$

for the dependence of τ_{exp} on the overinjection $\delta N/N_{\text{th}}$ is obtained from (1) and (2). With the BCS density of states N_{th} is approximated in the temperature range $T < 0.5 T_c$ by

$$N_{\text{th}} = N_0 [2\pi \Delta(T) k_B T]^{1/2} \exp[-\Delta(T)/k_B T], \quad (4)$$

where N_0 designates the density of electronic states of two spins per unit energy at the Fermi level.

Writing $x(0)$ for the relative change $\delta N(0)/N_{\text{th}}$ of quasiparticle concentration during injection we obtain from (1), (2) and (4) the resulting formula for N_0

$$N_0 = \frac{2I_0 \tau_{\text{exp},0} \exp[\Delta(T)/k_B T]}{[x^2(0) + 2x(0)] [2\pi \Delta(T) k_B T]^{1/2}}. \quad (5)$$

Thus, we need the absolute values of the measurable quantities T , Δ , I_0 , $\tau_{\text{exp},0}$ and $x(0)$ for an evaluation of N_0 from (5). These values can be obtained from a single symmetric superconducting tunneling junction $S_1|\text{oxide}|S_1$, i.e. $\Delta(T)$ and T from the current (i) – voltage (U) – characteristic and $x(0)$, $\tau_{\text{exp},0}$ from the time decay of an excess quasiparticle density.

For the following it is important that the thermal tunneling current i_{th} for junction voltages $k_B T/e < U < 2\Delta/e$ is directly proportional to N_{th} [18], whereas at the gap-voltage $2\Delta/e$ additional, nonthermally excited quasiparticles ($E \geq \Delta$) are injected into the tunneling volume V_{Tu} by breaking up Cooper-pairs. Accordingly we have

$$\delta N / N_{\text{th}} = \delta i / i_{\text{th}} \quad (6)$$

in the bias regime below $2\Delta/e$ and

$$I_0 = 2i_{\text{eff}}/eV_{Tu}, \quad (7)$$

for a sufficiently high current pulse of strength i_p driving the voltage of the junction, biased at $U_B < 2\Delta/e$, to $2\Delta/e$. Preconditions for the exactness of (6) are that δi and i_{th} are measured at the same bias and that the excess quasiparticles under stationary injection rate have the same energy distribution as the thermally excited quasiparticles N_{th} . $\tau_{\text{exp},0}$ is directly observed from the time decay of $\delta i(t)$ sufficiently long after switch-off of the injection pulse.

For obtaining the effective excitation current i_{eff} the thermal single-particle current i_{th} (at $U = 2\Delta/e$), the extra current due to the higher density of quasiparticles $\delta i(0)$, the leakage current i_L (at $U = 2\Delta/e$) and the so-called two-particle current i_{2P} [19] (at $U = 2\Delta/e$) – (or excess tunneling current at $U \geq \Delta/e$ due to contributions of the AC -Josephson-effect) – have to be subtracted from $i_p \pm i_B$, where the positive (negative) sign is valid if pulse current i_p and DC -current $i_B(U_B)$ have the same (opposite) direction. The total injection rate including i_{2P} as an excitation current is, however,

$$\begin{aligned} I_{0,\text{total}} &= 2i_{\text{eff}}/eV_{Tu} + i_{2P}/eV_{Tu} \\ &= 2\{i_p \pm i_B(U_B) - i_L(2\Delta/e) \\ &\quad - i_{\text{th}}(2\Delta/e)[x(0) + 1] - i_{2P}(2\Delta/e)/2\}/eV_{Tu}. \end{aligned} \quad (8)$$

A simultaneous measurement for i_p and $\delta i(0)$, i.e. $\delta i(0)/i_{\text{th}}$ is not possible. However, under certain conditions the nonlinear rate equations for disturbed quasiparticle and phonon systems [16] can be simplified and integrated and thus furnish a nonexponential decay function $\delta N(t)/N_{\text{th}} = x(t)$ after injection, so that, knowing the value of $\delta N/N_{\text{th}}$ for some time t , we can reextrapolate to $t=0$ and obtain the value $x(0)$ during the injection. Writing \dot{x} for dx/dt we derive

from the rate equations the exact expression

$$\bar{x} + \dot{x} \left\{ \frac{x+1}{\tau_R} + \frac{1}{\tau_{ph}} + \frac{1}{\tau_\gamma} \right\} + \frac{1}{2\tau_\gamma\tau_R} x(x+2) = 0. \quad (9)$$

τ_{ph} and τ_γ are the lifetimes of a gap-frequency phonon in the superconducting tunneling junction due to reabsorption and processes other than pair excitation respectively, i.e. phonon transmission across the junction surface boundaries and inelastic volume phonon interactions [20].

We substitute $\dot{x} = p$ and $\ddot{x} = p dp/dx$ and insert $\tau_{exp,0} = \tau_\gamma\tau_R(\tau_{ph}^{-1} + \tau_\gamma^{-1})$ [16] in Equation (9) and obtain

$$\frac{dp}{dx} + \frac{x+1}{\tau_R} + \frac{\tau_{exp,0}}{\tau_\gamma\tau_R} + \frac{1}{2\tau_\gamma\tau_R} \frac{x(x+2)}{p} = 0. \quad (10)$$

Neglecting the first and second term this equation can be integrated whence follows:

$$x(t) = 2\{[1 + 2/x(0)] \exp(t/\tau_{exp,0}) - 1\}^{-1} \quad (11)$$

or

$$\begin{aligned} x(0) &\equiv x(t=0) \\ &= 2\{[1 + 2/x(t)] \exp(-t/\tau_{exp,0}) - 1\}^{-1}, \end{aligned} \quad (12)$$

which is the value of $\delta N/N_{th}$ required in (5). A mean least-squares fit with Equation (11) on the experimental decay curves supplies values for $x(0)$ and $\tau_{exp,0}$. Comparing the terms of (9) by using the above approximate solution we find that within its validity range the relation

$$\tau_\gamma, \tau_{ph} \ll \tau_{exp,0}, \tau_R \quad (13)$$

is true for $x(0) \leq 1$. τ_γ can be adjusted by a proper choice of the junction thickness d and is generally in the order of the phonons' time of flight through the junction due to their efficient radiation into liquid helium [2], i.e. typically 10^{-10} s for d -values of the order of 10^3 \AA . The condition $\tau_{ph} \ll \tau_R$, $\tau_{exp,0}$ can be met by measuring at a sufficiently low temperature, since we obtain from detailed balance consideration of the thermal equilibrium [16, 17]

$$\begin{aligned} \frac{\tau_{ph}}{4\tau_R} &= \frac{N_{\omega,th}}{N_{th}} \\ &\simeq \frac{2\Delta^2(2c_l^3 + c_t^3)}{N_0(2\pi^5)^{1/2} c_l^3 c_t^3 \hbar^3} \left(\frac{Z^{1/2} + Z^{3/2} + Z^{5/2}/2}{\exp(1/Z)} \right), \end{aligned} \quad (14)$$

with $Z = k_B T/\Delta$; $N_{\omega,th}$ = total density of thermally excited 2Δ -phonons and c_l , c_t = longitudinal and transverse sound velocity. At $Z = 0.16$ ($T = 1.1$ K) we have $\tau_R/\tau_{ph} = 155$ for tin.

Furthermore,

$$\tau_{exp,0} = \tau_R(1 + \tau_\gamma/\tau_{ph}) \geq \tau_R. \quad (15)$$

An upper limit of $2 \cdot 10^{-10}$ s for τ_{ph} follows from measurements of thickness dependence of $\tau_{exp,0}$ [17]. Finally, an assumption implicit in the rate equations is that the quasiparticle diffusion length L_D is much greater than the junction film thicknesses. From measurements of residual film resistances we have determined the electron mean free path due to elastic scattering in the order of the film thicknesses and then we calculated $L_D \simeq 15 \mu$. Consequently, injected quasiparticles are homogeneously distributed by diffusive motion within the junction volume V_{Tu} before recombination takes place. We express this by a single value δN throughout V_{Tu} .

III. Experimental Realization and Data

The electronic measuring system for generating excess quasiparticles by current- and/or laser pulse excitation and recording the corresponding signal decays is shown in Figure 1 [12]. The injection current is supplied from a fast pulse generator

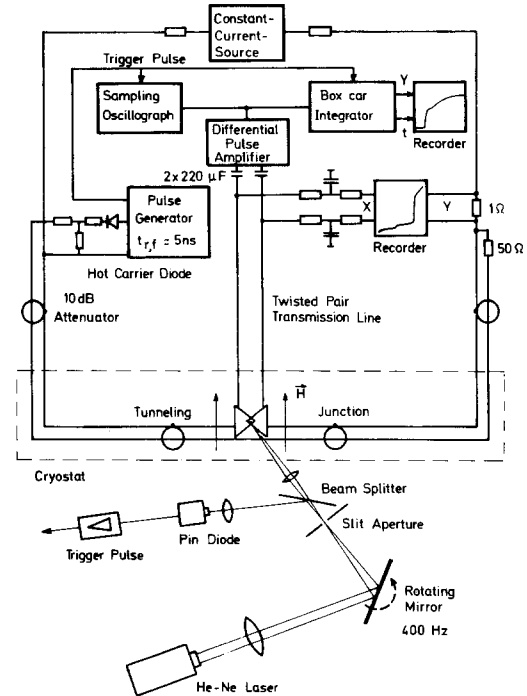


Fig. 1. Schematic diagram of the electrical circuitry for generating excess quasiparticle concentrations in a single superconducting tunnel junction $S_1|\text{oxide}|S_1$ by current and laser pulses and recording the signals as a function of time. The junction, acting simultaneously as generator and detector is operated at a constant DC-bias current. The blocking capacitors ($2 \times 220 \mu\text{F}$, tantalum) of low self-inductance are built in the signal circuit with negligible induction. To avoid any error, the current-voltage characteristics were recorded with open parallel current circuit (inner conductor of the coaxial cable)

($\tau_{r,f} \approx 4$ ns) connected via a hot carrier diode (Hewlett Packard, Type 5082-2810, $\tau = 100$ ps) to a $50\ \Omega$ -solid-jacketed coaxial cable (0.3 mm outer diameter), directly leading to the sample and then out of the cryostat. This coaxial transmission line is reflection-free terminated with a $50\ \Omega$ -resistor at room temperature. The current terminals of the junction are connected in series with both the ends of the interrupted outer conductor of this cable (see also Fig. 3). The influence of the asymptotic junction tunneling resistance R_∞ (typically in the $m\Omega$ -range) on the shape of the primary current pulse can be neglected. The hot carrier diode reduces the time constant of the current pulse and, moreover, suppresses any existing residual ripple. Thus, the switch-off time of the injection pulse is sufficiently well defined. This is important for an exact determination of $\chi(0)$. As to our junctions this switch-off decay time was about 10 ns. In addition, a small superimposed *DC*-current serves to bias the junction below $2\Delta/e$ after the switch-off of the current pulse (point 1 in Fig. 2). Since the junction is now operated at a constant *DC*-current, instead of the decaying excess thermal tunneling current $\delta i(t)$, a time dependent voltage $\delta U(t)$ is measured according to

$$\delta U(t) = -\delta i(t) (R_d \parallel R_L). \quad (16)$$

R_d , typically in the $1\ \Omega$ -range for the used junctions, is the dynamic resistance at the biasing point U_B and R_L the load resistance of the voltage measuring circuit.

The technique for generating sufficiently fast ($\tau_{r,f} < 100$ ns) light pulses is fundamentally simple and shown in the lower part of Figure 1. The light of a 15 mW-He-Ne laser is focused in the plane of a slit aperture after the reflection at a mirror. By rotating the mirror the reflected laser beam is mechanically chopped at the fixed aperture. The outgoing laser pulses with typical time constants of the leading and trailing edge and half-widths in the order of 50 ns are focused through the walls of a glass cryostat on the front of the junction. To avoid lattice heating effects in the quasiparticle photoexcitation experiments we used, according to [3], junctions prepared on sapphire substrates which were directly immersed into liquid helium II. Furthermore, although the quasiparticles are produced within the front junction layer in an optical penetration depth of a few hundred angströms, the excess quasiparticle concentration will be approximately uniform throughout the tunneling volume because of the large quasiparticle diffusion length and the indirect action of the recombination phonons.

A symmetrical, twisted pair transmission line (see also Fig. 3) connects directly the voltage terminals of the junctions with the input of a fast differential pulse

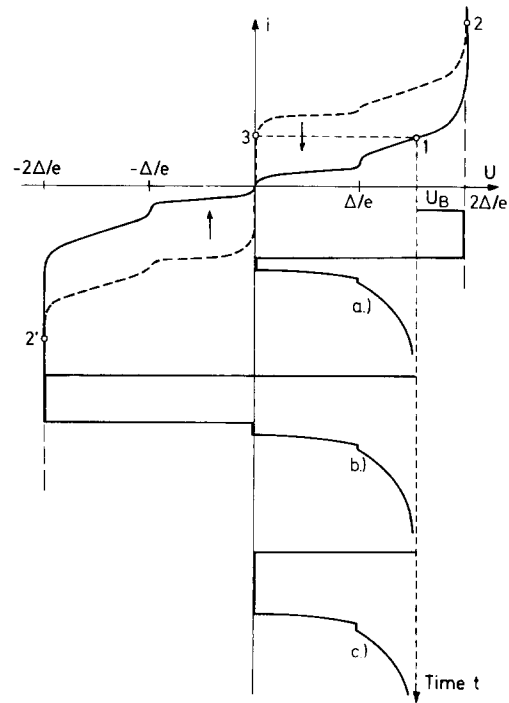


Fig. 2. Schematic drawing of the formation of the time dependent signals due to quasiparticle excitations by current (a, b) and laser pulses (c) taking account of the constant *DC*-current operation. Point 1 \triangleq bias voltage without injection, point 2 \triangleq biasing point shifted to the gap voltage due to the superimposed pulse current, point 3 \triangleq biasing point shifted to lower voltages due to the excess quasiparticle concentration after the switch-off time of the injection pulse

amplifier. The characteristic line impedance $Z \approx 100\ \Omega$ has been carefully matched to the input resistance $R_a = R_L = 2 \times 50\ \Omega$ of the amplifier and inductive geometries on the voltage side could be largely avoided. The amplified voltage signal is sampled and filtered in a PAR boxcar integrator (bandwidth 50 MHz, gate width minimum 10 ns) and then displayed by *X-Y*-recording as a function of time.

The time dependent shapes of the voltage signals to be expected with quasiparticle injection by current and laser pulses are schematically illustrated in Figure 2. The detailed shapes can be explained by the highly nonlinear *i-U*-characteristics in consideration of the constant *DC*-current operation and the quasiparticle density dependence of τ_{exp} [compare (3)]; e.g. the kink in the trailing edge of the voltage pulse corresponds to the current step at $U = \Delta/e$ in the tunneling characteristic. Generally speaking, a convex (concave) curvature of the characteristics as near to the bias level $U = 0$ ($U = \Delta/e$ and $2\Delta/e$) results in an increase (a decrease) of the real τ_{exp} .

Provided, however, that the dynamic resistance R_d is constant and independent of temperature in the operating region of the junction characteristic, the

voltage signal $\delta U(t)$ will be proportional to $\delta i(t)$ and thus to $\delta N(t)$. We, therefore, measured the voltage dependence of R_d for various temperatures and found out that R_d changed approximately to 1% in the voltage range $0.675 \text{ mV} \leq U_B \leq 0.825 \text{ mV}$ and within temperature intervals of 0.1 K for the used tin-junctions. Consequently, we have chosen this linear operating region with a bias voltage of $U_B = 0.8 \text{ mV} \approx 0.7 \Delta$. In order to avoid switching from the biasing point U_B to the zero voltage state due to the DC-Josephson effect, the junctions were operated in a parallel magnetic field of 10 to 25 Oe. In the linear regime τ_{exp} is directly obtained from the time decay of the voltage signal and is only a function of the momentary overinjection $\delta N(t)/N_{\text{th}}$. The pulse receiving system has been calibrated in terms of δi by means of small, simulated signal currents of known amplitude.

To determine the effective quasiparticle recombination times without any corrections over a wide range, the electronic time constant τ_{sys} of the receiving system must be small compared with $\tau_{\text{exp},0}$. By analyzing our circuit we obtained for instance $\tau_{\text{sys}} = 30 \text{ ns}$ at the biasing point $U_B = 0.8 \text{ mV}$ with $R_d \approx 2 \Omega$. As simulation experiments showed, this time constant is due to the time constant (25 ns) of the electronic measuring system and to the effective RC time of the junction

$$\tau_{Tu} = C_{Tu} R_d R_L / (R_L + R_d). \quad (17)$$

C_{Tu} , the capacitance of the tunnel junction, amounts to appr. 25 nF/mm^2 tunneling area. By using the parallel plate model for the junction capacitance and assuming 15 \AA for the barrier thickness, we obtained for the dielectric constant of the Sn-oxide a value of 4 in good agreement with other tunneling experiments carried out, however, at Pb-samples [21, 22]. For any necessary consideration of τ_{sys} the measured signal time constants $\tau_{\text{meas},0}$ were corrected according the formula

$$\tau_{\text{exp},0}^2 = \tau_{\text{meas},0}^2 - \tau_{\text{sys}}^2. \quad (18)$$

Figure 3 shows the sample configuration used in our experiments. The tunnel junction is formed between two superconducting films elsewhere insulated by an approximately 0.3μ thick SiO layer. Thus, the conductor loops in the region of the tunneling area can be made small; the nominal area is about $0.5 \times 0.6 \text{ mm}^2$. The inductive crosstalk from the high injection current pulse to the voltage terminals is, as an estimate showed, essentially determined by the ratio of the SiO-thickness to the width of the junction and amounts to less than 10^{-3} . At the narrow ends of the sample, the films form a pair of tines, respectively.

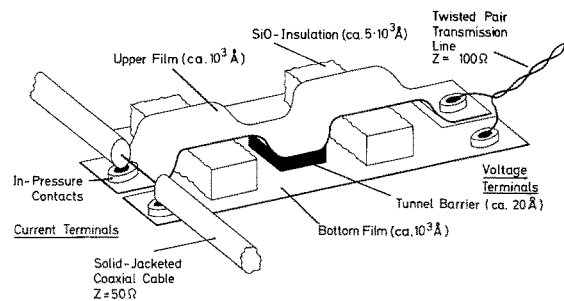


Fig. 3. Tunnel junction configuration for generating and detecting excess quasiparticles with low inductive crosstalk from the current pulse to the voltage terminals. The junction is vacuum-deposited on a sapphire or silicon substrate (not shown)

with an unshielded width of 0.1 mm. To minimize conductor loops at the leads, contact to the sample is made by pressing the current and voltage cable over the corresponding tines. Decay curves, experimentally obtained with this wide-band junction configuration, are free of disturbing, crosstalk-induced oscillations.

The samples were prepared in the conventional manner by using evaporation techniques. The films were vacuum deposited on a sapphire or silicon substrate under a residual gas pressure of 10^{-6} Torr and at a rate of 2 \AA/s to 200 \AA/s . Deposition in an oxygen background of about 10^{-5} Torr improves the junction quality, defined by the ratio $i_{2\Delta+}/i_{2\Delta-}$ of the maximal and minimal single particle current at $U = 2\Delta/e$. This current ratio is approximately equal to the resistance ratio R_d/R_∞ ; R_d below $U < 2\Delta/e$ is mainly determined by the leakage current. High R_d/R_∞ -values are important for the signal to noise ratio being proportional to R_d/R_∞ in this measuring method with high ohmic termination $R_L \gg R_d$. The junction thicknesses d were typically 4000 \AA to 5000 \AA . The tunneling barrier was formed by oxidizing the bottom film in an oxygen glow discharge for 5 min at a pressure of 0.08 Torr. We found out, that the junction quality doesn't depend on the sequence of the processes SiO-evaporation and Sn-oxidation. To prevent any condensation of SiO in the tunneling area, besides other precautions the distance between the corresponding evaporation mask and the bottom film was chosen as small as possible ($\leq 0.1 \text{ mm}$). The obtained low-ohmic samples (R_∞ in the $1 \text{ m}\Omega$ -range) with high quality ($i_{2\Delta+}/i_{2\Delta-} > 200$) indicated that the junction window wasn't coated with any disturbing SiO layer.

IV. Experimental Results and Conclusions

Figure 4 shows several time-dependent signal shapes, measured at low temperatures with the described technique in a slightly oxygen-doped (evaporation

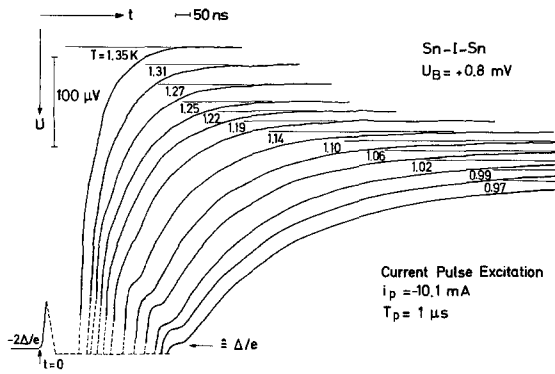


Fig. 4. Time-dependent voltage signal shapes in the linear regime of the i - U -characteristics at various temperatures. The curves are shifted vertically each other. The baselines correspond to U_B (biasing point 1 in Fig. 2)

pressure $8 \cdot 10^{-6}$ Torr O_2) tin junction under quasiparticle excitation by current pulses. The junction was biased in the linear range of the tunneling characteristics at $U_B = 0.8 \text{ mV} > \Delta/e$. Current pulses and bias current were opposite in polarity. In Figure 4 we have displayed with high sensitivity only that part of the total voltage signal decays ($2\Delta/e + U_B$), ranging from approximately Δ/e to the baseline ($U_B = 0.8 \text{ mV}$). The distinct kink in the various signal shapes is due to the relatively high excess DC-current $i_{2P} = 0.25 \text{ mA}$ at Δ/e in this sample (compare Fig. 2). To determine the switch-off time $t=0$ of the injection pulse, the signals were recorded with zero shift. The logarithmic diagrams of the displayed voltage amplitudes vs. time (not shown) result in straight lines at high temperatures ($> 1.10 \text{ K}$), while at low temperatures and high amplitudes (short times) the diagrams deviate from straight lines to shapes with greater slopes according to the effective overinjection. The slopes at sufficiently long times correspond to $\tau_{\text{exp},0}$ and agree with the time constants resulting from the fits of Equation (11) on the experimental decay curves. The differential time constants τ_{exp} , determined from the logarithmic diagrams at any overinjection are in agreement with Equation (3). Moreover, the signals shown in Figure 4 are independent of the pulse polarity; the signals obtained with negative or positive polarities and same effective injection rates differ less than 1%. Hence it follows an undisturbed symmetry of the used electronic measuring system.

The time constants $\tau_{\text{exp},0}$, measured from various runs with the same sample under current and laser-pulse excitations are plotted as a function of temperature in Figure 5 with the following results:

1) The exponential $\exp(\Delta(T)/k_B T)$ temperature dependence required by theory is verified by the $\tau_{\text{exp},0}$ values within $0.95 \text{ K} \leq T \leq 1.50 \text{ K}$; at these temperatures the relation $\Delta(T) \simeq \Delta(T=0) \equiv \Delta_0$ is valid for tin

with a measured maximal difference of 2%. The influence of $\tau_{\text{system}} = 30 \text{ ns}$ has been taken into account, compare Equation (18). Consequently, these time constants can be attributed to the effective quasiparticle recombination lifetimes in the tin tunnel junction.

2) The times measured below $T = 1.07 \text{ K}$ are corrected according to Equation (3) for the concentration dependence of τ , since additional quasiparticles are injected stationarily at the biasing point $U_B = 0.8 \text{ mV} > \Delta/e$ by the excess, “two-particle” current i_{2P} . This injection reduces $\tau_{\text{exp},0}$ to a saturation value at decreasing temperatures. The overinjection $\delta N(i_{2P})/N_{\text{th}}$ was determined from (1) and (2) and with the rate $I_0 = i_{2P}/eV_{Tu}$. With regard to this overinjection the lifetimes comply with the correct $\exp(\Delta_0/k_B T)$ -dependence at low temperatures too. Measurements on other tin samples with smaller i_{2P} showed a correspondingly reduced overinjection. At bias voltages $U_B < \Delta/e$ these lifetime corrections were not necessary. Thus, i_{2P} has to be considered as an injection current with the rate i_{2P}/eV_{Tu} .

3) $\tau_{\text{exp},0}$ is independent of quasiparticle excitations by current and laser pulses, respectively, within the accuracy in measurement of $\leq 10\%$.

4) Any magnetic flux trapped in the superconducting films, probably in form of quantized flux lines (vortices), effects an evident $\tau_{\text{exp},0}$ saturation at low temperatures (symbol (●) in Fig. 5). Responsible for this effect is a reduced reemission and reabsorption of 2Δ -phonons in the “vortices-doped” films with a strongly varying energy gap. Furthermore, the $i-U$ characteristics in the thermal tunneling range were shifted to higher currents with increasing voltage, e.g. by 8% of the total current at $U_B = 0.8 \text{ mV}$ and $T = 1 \text{ K}$. In Figure 5, the flux trapping was caused by the geomagnetic field acting perpendicularly on the tunneling area with a strength of only 0.2 G (measured with a Hall Sonde) when cooling down below T_c . In order to exclude largely these disturbing effects, the tunneling barrier was oriented parallel to the geomagnetic field.

5) Over the temperature range of our measurements, the effective quasiparticle recombination lifetime in the 4200 \AA thick superconducting tin tunnel junction on a sapphire substrate immersed in He II is

$$\tau_{\text{exp},0} = \tau_0 \frac{T^{-1/2}}{[K^{-1/2}]} \exp(\Delta_0/k_B T), \quad (19)$$

with $\tau_0 \simeq 0.6 \text{ ns}$. This value is compared with theory based on a ray-acoustic 2Δ -phonon trapping lifetime model [17] showing a linear thickness dependence of $\tau_{\text{exp},0}$ for the relations $d > A_w$ and $A_v \gg A_w$ between the mean free paths of 2Δ -phonons due to reabsorption (A_w) and bulk loss processes (A_v); $A_w \leq 700 \text{ \AA}$ in

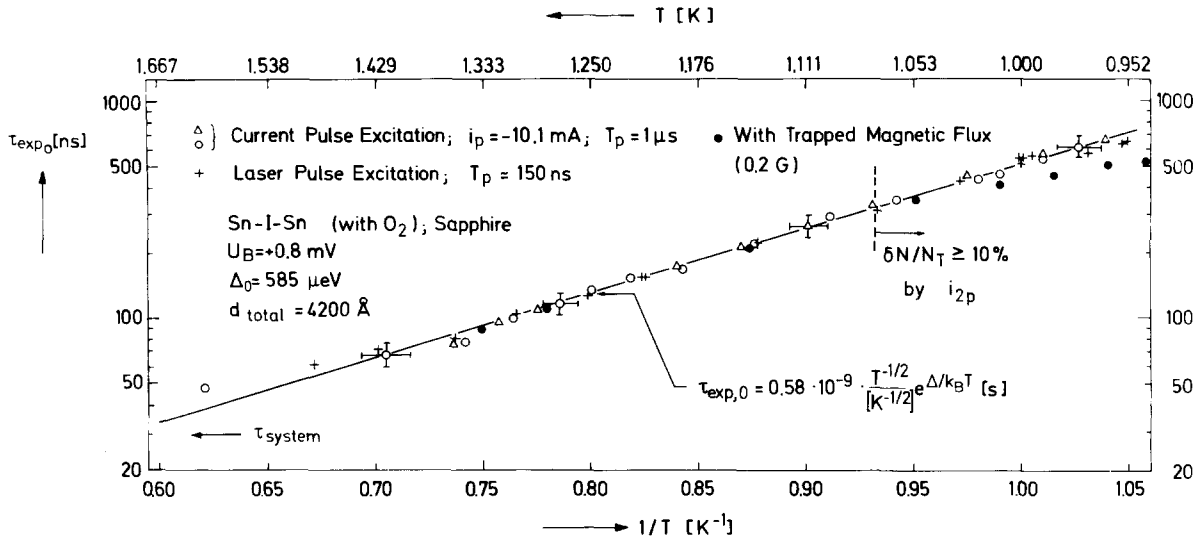


Fig. 5. Experimental quasiparticle recombination lifetimes $\tau_{\text{exp},0}$ versus $1/T$ for a superconducting tin-tunnel junction immersed in superfluid He. Overinjection by the “two-particle current” at the biasing point $U_B = 0.8 \text{ mV} > \Delta/e$ has been taken into account

Sn [17]:

$$\tau_{\text{exp},0} = d \frac{N_{\text{th}}}{N_{\omega,\text{th}}} \left(\frac{1}{c_t^3} + \frac{1}{2c_l^3} \right) / \left(\frac{\bar{T}_t}{c_t^2} + \frac{\bar{T}_l}{2c_l^2} \right), \quad (20)$$

where $\bar{T}_{t,l} = \bar{T}_{t,l;F/S} + \bar{T}_{t,l;F/He}$ the total average phonon transmission at the boundaries film (F)/substrate (S) and film/helium with

$$\bar{T}_{t,l} = \int_0^{\pi/2} T_{t,l}(\theta) \sin 2\theta d\theta, \quad (21)$$

the average value of the angle and polarization dependent phonon transmission coefficient $T_{t,l}(\theta)$. As the transverse phonon contribution to the transmission across the junction boundaries is dominant (appr. 85%), it follows from (20)

$$\tau_{\text{exp},0} = d \frac{N_{\text{th}}}{N_{\omega,\text{th}}} / c (\bar{T}_{F/S} + \bar{T}_{F/He}). \quad (22)$$

With the experimental data for N_0 and Δ_0 listed in Table 1 as well as the transverse sound velocity $c = 1.67 \cdot 10^5 \text{ [cm/s]}$ [17] and the relation $c_t \approx 2c_l$, we obtain from (14) in good approximation:

$$\frac{N_{\text{th}}}{N_{\omega,\text{th}}} = 1.56 \frac{T^{-1/2}}{[K^{-1/2}]} \exp(\Delta_0/k_B T) \quad (23)$$

by taking into account only the $Z^{1/2}$ term. Applying Equation (22) to the specified data and using $\bar{T}_{\text{Sn}/\text{Al}_2\text{O}_3} = 0.07$ (from acoustic mismatch model) [17] and $\bar{T}_{\text{Sn}/\text{He}} = 0.32$ (from τ_{exp} -measurements in vacuum and He) [2] or $\bar{T}_{\text{Sn}/\text{He}} = 0.5$ (from Kapitza resistance) (Ref. 8 in [5]) results in

$$\tau_0 = 1.0 \text{ ns or } 0.7 \text{ ns, respectively.} \quad (24)$$

In order to check this result we also measured the lifetimes in oxygen-free Sn-junctions (evaporated on sapphire in a vacuum of a residual pressure of

10^{-6} Torr), the other parameters, as for instance the contact to liquid He, being unchanged. Within the measurement accuracy there was no deviation compared to those in Figure 5. Therefore, the difference between the experimental (19) and calculated result (24) may not be ascribed to volume losses of 2Δ -phonons in the oxygen-doped tin samples or an enhanced 2Δ -phonon escape by diffuse phonon scattering at the junction boundaries. This is consistent with the data in Table 1 obtained for O_2 -free and O_2 -doped tin films. In view of the uncertainty involved in determining $\bar{T}_{\text{Sn}/\text{He}}$ the agreement between experiment and calculation seems to be good.

Experimental lifetime values in tin samples, evaporated, however, on silicon substrates turned out to be lower by approximately 10%. This reduction is within the measuring error, however can be fully accounted for the changed phonon transmission at the boundary tin/substrate.

Finally we have calculated a mean value N_0 for the density of electronic states at the Fermi level (presented in Table 1 with its plus–minus standard deviation) from Equation (5) and with data of the analyzed measurements on three superconducting tunnel junctions at then different temperatures in each case. The maximum scatter of the single N_0 values relating to the mean value amounts to 10%. From Equation (5) we estimated a maximal uncertainty for N_0 of 30% to 40% corresponding to the main sources of error: $\tau_{\text{exp},0} (\pm 5\%)$, $V_{Tn} (\pm 5\%)$, $x(0) (\pm 10\%)$ and $\Delta (\pm 2\%)$; the last-quoted results in an error of 12% in $\exp(\Delta/k_B T)$ at $\Delta = 6k_B T$. The presented value for N_0 from our direct quasiparticle lifetime experiments on thin, superconducting tin films of polycrystalline structure is to be compared

Table 1. Some characteristic parameters of tin films evaporated in a vacuum at a residual pressure of 10^{-6} Torr and/or a pressure of $8 \cdot 10^{-6}$ Torr oxygen. $\rho_{4,2}$ = resistivity measured at $T=4.2$ K, R = film resistance

Δ_0 [μeV]	T_c [K]	$2\Delta_0/k_B T_c$	δT_c [K]	d [\AA]	$\rho_{4,2}$ [$10^{-7}\Omega\text{cm}$]	$l_{4,2}$ [\AA]	ξ [\AA]	$R_{4,2}/R_{300}$	N_0^{tt} [$10^{22}\text{eV}^{-1}\text{cm}^{-3}$]
585	3.76	3.61	0.08	2000	6.2	1700	2000 at 1K	0.053	2.73 ± 0.03
For comparison: bulk value $N_0^{tt} = 2.77 \cdot 10^{22} \text{eV}^{-1} \text{cm}^{-3}$ from specific-heat measurements									

with the value determined from conventional measurements [23,24] of the electronic specific heat on ordinary bulk tin. The electronic heat capacity coefficient is proportional to the band-structure density of states N_0^b times the enhancement factor $(1 + \lambda)$ due to the electron-phonon coupling λ [25].

As Table 1 shows, there is an excellent agreement between the film and bulk values N_0 within the given limits of error. This result is compatible with other film properties we have also measured and included in Table 1. Thus, T_c (film) is only enhanced to 1% compared to T_c (bulk); for comparison we evaluated from the BCS equation for T_c an enhancement of T_c of 43% by varying exclusively N_0 to 10%. Moreover, the coherence length ξ at $T=1$ K is of the order of the film thickness d and the mean free path $l_{4,2}$ (at 4.2 K) of electrons due to elastic scattering at defects, internal and external film boundaries; $l_{4,2}$ gives approximately a lower limiting value for the average grain size \bar{D} in our tin films.

After testing the reliability of the described method on superconducting tin tunnel junctions we have also applied it to aluminium and lead samples with different aspects in each case [12]. By experiments on Al we wanted to answer the question, whether the measured T_c enhancement of oxygen-perturbed, granular Al films may be caused by a change in N_0 . Furthermore, we checked whether the effective quasiparticle recombination lifetimes are changed with increasing perturbations of the film structure. Measurements on high-quality lead junctions under vacuum conditions were made in order to draw conclusions from the effect of inelastic 2Δ -phonon bulk loss processes on the value of $\tau_{\text{exp},0}$. A detailed description of these experiments will be published separately.

References

1. Eisenmenger, W.: in: Tunneling Phenomena in Solids, E. Burstein and S. Lundquist (eds.), p. 371. New York: Plenum Press 1969

2. Trumpp, H.J., Lassmann, K., Eisenmenger, W.: Phys. Lett **41**A, 431 (1972)
3. Parker, W.H., Williams, W.D.: Phys. Rev. Lett. **29**, 924 (1972)
4. Sai-Halasz, G.A., Chi, C.C., Denenstein, A., Langenberg, D.N.: Phys. Rev. Lett. **33**, 215 (1974)
5. Parker, W.H.: Sol. State Comm. **15**, 1003 (1974)
6. Hu, P., Dynes, R.C., Narayanamurti, V.: Phys. Rev. **B10**, 2786 (1974)
7. Ginsberg, D.M.: Phys. Rev. Lett. **8**, 204 (1962)
8. Müller, B.I., Dayem, A.H.: Phys. Rev. Lett. **18**, 1000 (1967)
9. Levine, J.L., Hsieh, S.Y.: Phys. Rev. Lett. **20**, 994 (1968)
10. Gray, K.E., Long, A.R., Adkins, C.J.: Phil. Mag. **20**, 273 (1969)
11. Sonnenberg, K.H.: Diplomarbeit, Universität Göttingen (1970), unpublished
12. Epperlein, P.W.: Thesis, Universität Stuttgart (1977). Quasiparticle recombination lifetime measurements on a single superconducting tunnel junction by current pulse excitation were made for the first time by [10] and [11]. The requirements for the pulse electronics, however, increase in the sequence of experiments on Al [10,12], Sn [11,12] and Pb [12] because of the corresponding, decreasing lifetimes.
13. Gray, K.E.: J. Phys. F (Metal Phys.) **1**, 290 (1971)
14. Long, A.R.: J. Phys. F (Metal Phys.) **3**, 2040 (1973)
15. Smith, L.N., Mochel, J.M.: Phys. Rev. Lett. **35**, 1597 (1975)
16. Rothwarf, A., Taylor, B.N.: Phys. Rev. Lett. **19**, 27 (1967)
17. Eisenmenger, W., Lassmann, K., Trumpp, H.J., Krauss, R.: Appl. Phys. **11**, 307 (1976); Appl. Phys. **12**, 163 (1977)
18. Eisenmenger, W.: in: Physical Acoustics Vol. XII, 1976 (Eds. W.P. Mason and R. Thurston)
19. Schrieffer, J.R., Wilkins, J.W.: Phys. Rev. Lett. **10**, 17 (1963)
20. Schuller, I., Gray, K.E.: Phys. Rev. **B12**, 2629 (1975)
21. Zappe, H.H., Grebe, K.R.: J. Appl. Phys. **44**, 865 (1973)
22. Scott, W.C.: Appl. Phys. Lett. **17**, 166 (1970)
23. Kittel, C.: Introduction to Solid State Physics, IIIrd Edition. New York: J. Wiley & Sons 1967
24. Gopal, E.S.R.: Specific Heats of Low Temperatures, London: Plenum Press 1966. K. Mendelssohn and K.D. Timmerhaus (eds.) The bulk value for N_0 listed in Table 1 was evaluated from the relation $N_0 = 3\gamma/\pi^2 k_B^2$ with an average electronic specific heat coefficient γ , obtained from [23] and [24].
25. McMillan, W.L.: Phys. Rev. **167**, 331 (1968)

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