

Response of superconducting films to a periodic optical irradiation

N. Perrin

Groupe de Physique des Solides de l'École Normale Supérieure, Laboratoire associé au Centre National de la Recherche Scientifique, 24 rue Lhomond, F-75231 Paris Cedex 05, France

C. Vanneste

Laboratoire de Physique de la Matière Condensée, Laboratoire associé au Centre National de la Recherche Scientifique, Université de Nice, F-06034 Nice Cedex, France
(Received 12 July 1982; revised manuscript received 20 December 1982)

We present a theoretical study of the quantitative response of a superconducting film driven out of equilibrium by modulated laser irradiation. A numerical solution of the time-dependent coupled equations is performed, the quasiparticle distribution being represented by a Fermi-Dirac distribution with an effective temperature T^* . The time evolution of a typical Sn film is first studied at a drive frequency $f = 1$ GHz. The film response is then analyzed as a function of the drive frequency. It is shown that, in Sn, it is limited by the energy transfer from the quasiparticles to the phonon system. Although the film response depends on the phonon escape time and on the intrinsic times of the quasiparticle-phonon system, none of these times is shown to determine alone the time behavior. The characteristic times of the response also depend on the specific-heat ratios of the phonon and quasiparticle systems. Results are presented for the modulation depths and phase shifts of the quasiparticle temperature and of the power transferred to the phonon for different specific-heat ratios, intrinsic times, and phonon escape times. A comparison with the results obtained from a simple analytic model is done. We have also briefly discussed the situation for some other superconducting materials.

I. INTRODUCTION

Since the initial historical experience by Testardi,¹ optical irradiation has frequently been used to drive a superconductor in an out-of-equilibrium state. The aim of most of the experiments, performed either in the steady-state regime or in the pulsed regime (with microsecond-to-nanosecond laser pulses), was to reach the microscopic behavior of nonequilibrium superconductors through tests of quasiparticle-dynamics models,¹⁻³ determination of the distribution functions of the excitations,⁴⁻⁷ study of intermediate resistive states,⁸⁻¹¹ and determination of the intrinsic relaxation times.^{3,8,12-16} During the last years, particular interest has been devoted to the superconductor response to picosecond laser pulses.¹⁷⁻¹⁹ Moreover, some workers have considered periodically irradiated superconductors.^{20,21} From recent experimental work, it has been concluded that the response is limited in the high-excitation-frequency range by the escape time τ_{es} of the phonons out of the illuminated region,¹⁷ even when the laser-pulse duration is comparable to the electron-phonon interaction time.¹⁹ This seems to agree with the qualitative predictions which have been deduced²¹ from previous steady-state results.^{22,23} To get rid of this rather long limiting time ($\tau_{es} \simeq 10^{-9}$ sec) some experimentalists have proposed using variable-thickness bridges, where the quasiparticle diffusion out of the illuminated region becomes important.¹⁸ They expect a transient response time determined by the pair-breaking time for an efficient quasiparticle diffusion. However, these qualitative predictions are deduced from an analysis based on the simple Rothwarf-Taylor equations.²⁴

In this paper, we analyze the quantitative response of

the quasiparticle-phonon system through approximations of the complicated quasiparticle and phonon-collision integrals which are not so crude as in the Rothwarf-Taylor model. We study the behavior of a superconducting film perturbed by a periodic laser drive. We are particularly interested in the dynamics of the quasiparticle population, which is the relevant quantity in the determination of the superconducting parameters such as the order parameter Δ , which we consider below, and the critical current in a junction. The role of the phonons, which, in the steady state, is to transfer the input power from the quasiparticle system to the substrate, is considered. Indeed, the phonons provide an energy releasing mechanism through the substrate-film contact and a heat reservoir, which may both be determinant in the time behavior of the whole system.

We consider different frequencies in the GHz range; the excitation periods are thus comparable to the electron-phonon interaction times and to the phonon escape time in the Sn film studied. This is not the case for strong coupling superconductors such as Pb and Nb which electron-phonon interaction times are small; indeed, the electron-phonon relaxation time is about 10 psec (Pb) and the phonon pair-breaking relaxation time can be as short as 10 psec (Pb) or smaller (a few picoseconds in Nb),^{18,25} and then far shorter than the phonon escape time.

Moreover, the quasiparticle system acts, like the phonon system, as a heat reservoir, and we are going to see below that, depending on the relative specific heat of these two reservoirs, quite different time behaviors of the superconducting film are expected. Therefore, we study here the influence of characteristic quantities such as the intrinsic relaxation times of the excitations and the specific heat on

the film response. The results obtained allow us to show which response is expected from superconducting films such as Sn or In films under periodic laser irradiation and to foresee the time behavior of Pb or Nb films, in the same conditions of irradiation.

A detailed study of the coupled dynamics of quasiparticles, pairs, and phonons is performed here through the numerical solution of the relevant coupled equations.

The film is assumed to be uniformly illuminated by the laser light. The quasiparticles absorb the optical power; their coupling with the phonons leads to phonon deviations from equilibrium. We thus study the quasiparticle-phonon energy exchanges. Amplitudes and phase shifts are considered for the latter as well as for the quasiparticle temperature T^* .

The calculation model and the coupled equations for quasiparticles, pairs, and phonons are presented in Sec. II. In Sec. III, the numerical method used to solve these equations is given and the results are presented in Sec. IV. In Sec. V, we compare our numerical results with the analytic results deduced from a simple macroscopic model for both phonons and quasiparticles.

II. FORMULATION (REF. 23)

The quasiparticle distribution function $f(E)$ is taken to be a spatially uniform Fermi-Dirac function at an effective time-dependent temperature $T^*(t)$, with a chemical potential $\mu=0$ as in the BCS theory.²⁶ These two assumptions require both a very short time for the thermalization of the quasiparticles¹ at T^* and a time for recombination shorter than the time for quasiparticles to thermalize with respect to the low-energy phonons. This latter condition is indeed satisfied in thermal equilibrium in a large region of energies E for $T^* > 0.5T_c$ (Refs. 1, 25, and 27–29) and remains valid for nonequilibrium superconductors since the recombination time decreases with excess quasiparticles. The use of a single temperature T^* to characterize the quasiparticle distribution is of course an approximation, the full exact calculation requiring the solution of the kinetic equations for both quasiparticles and phonons. However, for laser drive, the initially generated excitations are broadly distributed and a T^* Fermi-Dirac distribution is the more closely approached as the phonon coupling with the substrate is weak.²² Moreover, it has been shown experimentally that the measured quasiparticle distribution for optically illuminated films is well described by a T^* distribution.^{4,5,7} The T^* Fermi-Dirac distribution ap-

proximation therefore remains a good approximation for laser-optical irradiation in the case of a mean coupling between the phonons and the substrate. It retains the essential physics of our problem but considerably simplifies the mathematical analysis.

The time-dependent superconducting gap Δ is determined by the instantaneous nonequilibrium distribution function $f(E)$ through the BCS equation,

$$\Delta/\Delta_0 = \exp \left[- \int_{-\infty}^{+\infty} d\epsilon f(E)/E \right], \quad (1)$$

which we assume to hold instantaneously; Δ_0 is the zero-temperature gap and $\epsilon = \pm(E^2 - \Delta^2)^{1/2}$.

Therefore, the energy balance for the quasiparticle gives

$$C_e \frac{dT^*}{dt} = P_i(t) - P_N(t). \quad (2)$$

$P_i(t)$ is the input power, $P_N(t)$ is the power lost by the quasiparticles and transferred to the phonons, and C_e is the quasiparticle specific heat. In order to avoid its complete calculation from the BCS equation, we use the following expression³⁰:

$$C_e = a\gamma T_c \exp(-bT_c/T^*), \quad (3)$$

where γ is the ratio of the normal-metal specific heat to the temperature. Equation (3) is quite a good approximation for Sn with $a \simeq 9$ and $b \simeq 1.5$.³⁰ This approximation introduces no important quantitative error in the range of temperatures considered below.

The power received by the phonons from the quasiparticles is given by

$$\begin{aligned} P_N(t) &= \sum_{\vec{q}, \alpha} \hbar\omega \frac{\bar{N}(T^*) - N_\alpha}{\tau_{P\alpha}} \\ &= \sum_{\vec{q}, \alpha} \hbar\omega \left[\frac{\partial N_\alpha}{\partial t} \right]_e. \end{aligned} \quad (4)$$

$\bar{N}(T^*)$ is the Planck distribution at T^* and N_α is the phonon distribution in the mode α , assumed to be spatially uniform.³¹ The phonon relaxation time τ_P is given by

$$\tau_{P\alpha}^{-1} = (\tau_{PB}^{-1} + \tau_{PS}^{-1})_\alpha \equiv \tau_P^{-1}, \quad (5)$$

the phonon pair-breaking and -scattering relaxation times, τ_{PB} and τ_{PS} , being, respectively, given by²³

$$\tau_{PB}^{-1} = \begin{cases} C \int_{\Delta}^{\hbar\omega - \Delta} dE \rho(E) \rho(\hbar\omega - E) \left[1 + \frac{\Delta^2}{E(\hbar\omega - E)} \right] [1 - f(\hbar\omega - E) - f(E)] & \text{for } \hbar\omega > 2\Delta \\ 0 & \text{for } \hbar\omega \leq 2\Delta \end{cases} \quad (6)$$

and

$$\tau_{PS}^{-1} = 2C \int_{\Delta}^{\infty} dE \rho(E) \rho(E + \hbar\omega) \left[1 - \frac{\Delta^2}{E(E + \hbar\omega)} \right] [f(E) - f(\hbar\omega + E)]. \quad (7)$$

These quantities are time dependent through T^* and Δ . The constant C , which partly results from the electron-phonon matrix element, writes, in the deformation-potential approach,

$$C = \frac{(mE_\alpha)^2}{2\pi\hbar^4\rho_D u_\alpha}.$$

m is the electron mass, ρ_D , E_α , and u_α are, respectively,

the density of the crystal, the deformation-potential constant, and the sound velocity in the α mode.

The phonon distribution N_α is computed from the phonon Boltzmann equation

$$\frac{\partial N_\alpha}{\partial t} = \left[\frac{\partial N_\alpha}{\partial t} \right]_{\text{es}} + \left[\frac{\partial N_\alpha}{\partial t} \right]_e, \quad (8)$$

where $(\partial N_\alpha / \partial t)_e$ gives the effect of the quasiparticles on the evolution of the phonon distribution whereas the rate of change of N_α due to phonons escaping from the film can be written phenomenologically as follows:

$$\left[\frac{\partial N_\alpha}{\partial t} \right]_{\text{es}} = - \frac{N_\alpha - \bar{N}(T_b)}{\tau_{\text{es}}}. \quad (9)$$

The time τ_{es} represents the time for the phonons to thermalize to the bath temperature at T_b ; it is of the order of the phonon transit time d/u : $\tau_{\text{es}} = \eta d/u$ (d is the film thickness, u is the mean sound velocity, and η is a constant, determined by the phonon transmittivity).

III. NUMERICAL METHOD

The time variations of the phonon distribution function and of the quasiparticle effective temperature T^* can be determined from Eqs. (1)–(9).

We assume that the system is initially in a stationary state characterized by the values T_S^* and $N_{\alpha S}$, respectively, of the quasiparticle temperature and of the phonon distribution. Then, the modulation of the input power $P_i(t)$ is applied at $t_0=0$; we choose a sinusoidal form,

$$P_i(t) = P_S(1 + \sin \omega_i t). \quad (10)$$

P_S is thus the input power corresponding to the initial stationary state.

Therefore, we first perform a numerical evaluation of P_S along with the corresponding value of $N_{\alpha S}$ for a given T_S^* :

$$P_S = \sum_{\vec{q}, \alpha} \hbar \omega [\bar{N}(T_S^*) - N_{\alpha S}] / \tau_{P\alpha} = P_{NS}, \quad (11)$$

where P_{NS} is the power transferred to the phonons in the initial stationary state. Then, a small time interval Δt is considered. During this time, T^* and N_α undergo small variations, δT^* and δN_α , which we calculate from Eqs. (2) and (8). Having thus determined the values of T^* , we determine Δ and P_N from Eqs. (1) and (4), and we continue to the next interval. The time interval Δt is chosen such that

$$\Delta t < \min \left[\frac{f^{-1}}{20}, \tilde{\tau} \right], \quad (12)$$

where $\tilde{\tau}$ is a time smaller than all the relaxation times in the system.

IV. RESULTS

We choose a Sn film with $\Delta_0 = 0.561$ meV (6.5 K), $\rho_D = 7$ g cm $^{-3}$, and $m = m_0$, the free-electron mass. The bath temperature is $T_b = 1.5$ K.

Both transverse and longitudinal phonons are considered. A deformation-potential constant $E_\alpha = E_1 = \frac{2}{3} E_F = 6.89$ eV is taken for the coupling of the electrons

with the longitudinal phonons. In tin, the coupling of the electrons with the transverse phonons is important owing to the departure from the free-electron model. However, it has been shown that this real-metal effect can be considered in a deformation-potential approximation with a deformation-potential constant $E_2 = 2.5$ eV for both of the transverse modes.³²

Different phonon escape times, $\tau_{\text{es}} = 0.1, 0.3$ nsec, are considered. For excitation frequencies larger than the characteristic frequencies of the system, the whole system is expected to be quite insensitive to the modulated excitation. Therefore, we choose excitation frequencies ω_i smaller than 10 GHz; this frequency just corresponds to the condition $\omega_i \tau \gg 1$ in our work.

Under these conditions, the results obtained are shown in Figs. 1–9. A fixed frequency $f = 1$ GHz is first considered (Sec. IV A) for which the time behavior of the quasiparticle temperature and of the power transferred to the phonons and the effect of the phonon and quasiparticle specific heat are analyzed. Then, the frequency is varied (Sec. IV B).

A. Time behavior at a fixed frequency; importance of the specific heat

In Figs. 1 and 2, a typical time behavior is shown of the quasiparticle effective temperature T^* , of the gap parameter Δ , and of the power $P_N(t)$ received by the phonons from the quasiparticles, for a phonon escape time $\tau_{\text{es}} = 0.3$ nsec. The initial quasiparticle temperature and input power are, respectively, $T_S^* = 2.98$ K and $P_S = 14.1 \times 10^4$ W cm $^{-3}$.

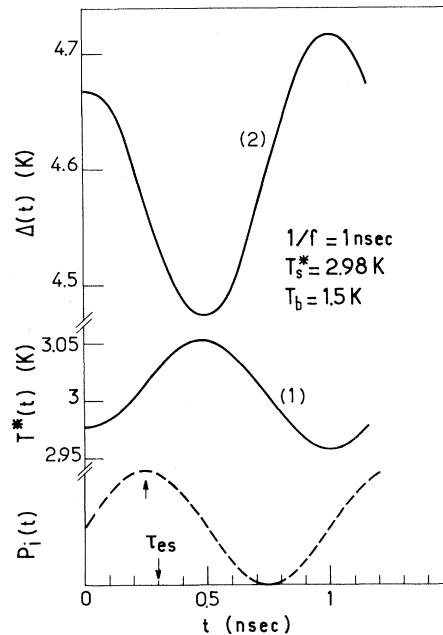


FIG. 1. Quasiparticle temperature $T^*(t)$, gap parameter $\Delta(t)$, and input power $P_i(t)$ (arbitrary units) vs time t , for a drive period $f^{-1} = 1$ nsec and a transverse and longitudinal phonon escape time $\tau_{\text{es}} = 0.3$ nsec.

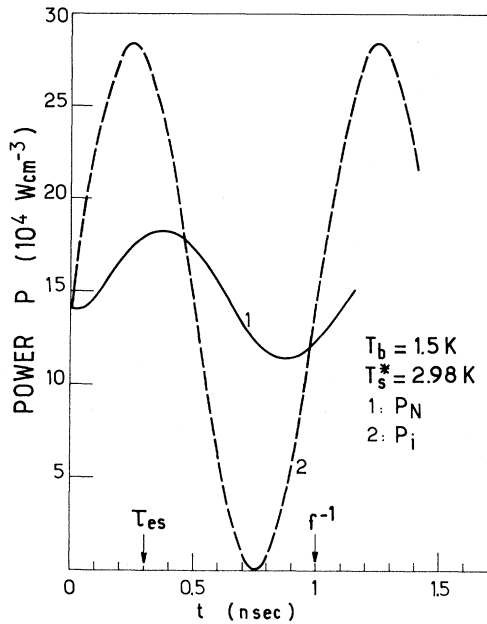


FIG. 2. Input power $P_i(t)$ and power $P_N(t)$ transferred from the quasiparticles to the phonons vs time t for a drive period $f^{-1}=1$ nsec and an escape time $\tau_{es}=0.3$ nsec for both transverse and longitudinal phonons.

The quasiparticle-temperature modulation depth,

$$t^*/(T_S^* - T_b) = (T_{\max}^* - T_S^*) / (T_S^* - T_b),$$

is of about 5% which is rather small [curve 1 in Fig. 1] though the frequency f is smaller than most of the characteristic frequencies in the system as can be seen in Table I. This can be understood [Eq. (2)] from the large value of the quasiparticle specific heat C_e , at the temperatures considered, compared to the phonon specific heat C_p ($C_e/C_p \approx 1.5$ with $C_e \approx 430 \text{ J m}^{-3} \text{ K}^{-1}$ and $C_p \approx 300 \text{ J m}^{-3} \text{ K}^{-1}$) as it will be seen below. Therefore, a rather small modulation of the gap parameter Δ ($\approx 4\%$) is obtained [curve (2) in Fig. 1] although the initial temperature $T_S^* = 0.8T_c$ has been chosen in a region where the variations of Δ with T^* become important according to the BCS theory.

The quasiparticle temperature T^* [curve (1) in Fig. 1 (Ref. 33)] is seen to be nearly in quadrature with the input power $P_i(t)$ (dashed curve in Fig. 1), the phase shift between T^* and $P_i(t)$ being $\varphi_e \approx 83^\circ$. This is due to the rather small role of P_N [Eq. (2)] in the case considered.

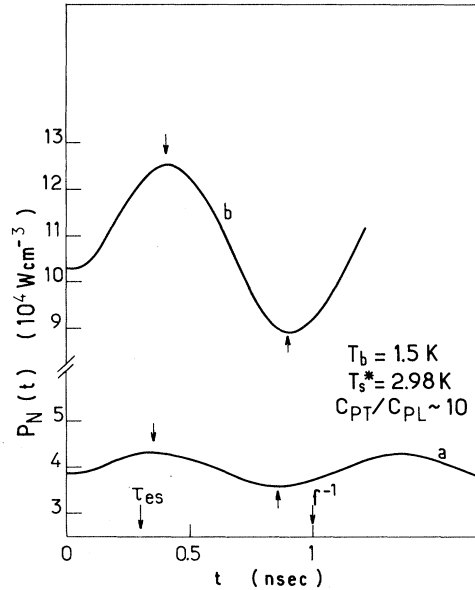


FIG. 3. Power transferred from the quasiparticles to the phonons $P_N(t)$ vs time, taking account of the longitudinal phonons only [A system; curve (a)], or of the transverse phonons only [B system; curve (b)] for a drive period $f^{-1}=1$ nsec. The phonon escape time is $\tau_{es}=0.3$ nsec.

From Fig. 2 it is seen that the amplitude $P_N = P_{N\max} - P_{NS}$ of the power transferred from the quasiparticles to the phonons [curve 1] is far smaller than the amplitude $p_i = P_{i\max} - P_i$ of the input power $P_i(t)$ [dashed curve (2)], their ratio being $p_N/p_i \approx 0.3$. The variations of $P_N(t)$ are complicated since P_N depends on T^* in a non-linear way essentially through $\bar{N}(T^*)$ [Eq. (4)], but it also depends on the heating of the phonon system through the phonon distribution N_α [Eq. (4)], and on the rate at which the phonons absorb energy from the quasiparticles which is characterized by the phonon relaxation time τ_p [Ref. 34, Eq. (4)], these factors being all interdependent.

The importance of the phonon specific heat in the amplitude response of $P_N(t)$ is seen in Fig. 3 where we have considered two different systems: the first (A system) includes pairs, quasiparticles, and longitudinal phonons only; the second (B system) includes pairs, quasiparticles, and transverse phonons only; the corresponding calculated time behavior of $P_N(t)$ is represented, respectively, by curves (a) and (b) for an initial quasiparticle temperature $T_S^* = 2.98 \text{ K}$ and a phonon escape time $\tau_{es} = 0.3$ nsec. The

TABLE I. t_0 is the initial time, at which the modulation of the input power P_i is applied; the times t_1 and t_2 correspond to the maxima of $P_i(t)$ and $T^*(t)$, respectively. $\tau_{PBL(T)}$ and $\tau_{PSL(T)}$ are, respectively, the phonon pair-breaking and -scattering relaxation times for 2Δ energy phonons; τ_p is the phonon relaxation time $\tau_p^{-1} = \tau_{PS}^{-1} + \tau_{PB}^{-1}$. The indices L and T stand, respectively, for the longitudinal and transverse phonons which are *simultaneously* considered here, with a phonon escape time $\tau_{es} = 0.3$ nsec and a drive frequency $f = 1 \text{ GHz}$. The times are expressed in nsec.

	τ_{PBL}	τ_{PSL}	τ_{PL}	τ_{PBT}	τ_{PST}	τ_{PT}	T^* (K)
t_0	0.135	0.833	0.116	0.601	3.715	0.517	2.9776
t_1	0.141	0.800	0.120	0.627	3.556	0.533	3.015
t_2	0.147	0.763	0.123	0.656	3.41	0.550	3.046

TABLE II. Phonon escape time τ_{es} , mean phonon relaxation time $\tau_P(2\Delta)$, and the corresponding characteristic times, τ_1 and τ_2 , of the quasiparticle-phonon system; C_e/C_P is the ratio of the quasiparticle and phonon specific heats. P_S is the initial input power corresponding to the same initial quasiparticle temperature $T_S^*=2.98$ K in all the cases considered. The (A1) and (A2) [B] cases correspond to longitudinal (transverse) phonons only being taken into account. The times are expressed in nsec, and $\tau_\varphi^{-1} = \tau_{es}^{-1} + \tau_P^{-1}$.

		τ_{es}	τ_P	τ_φ	τ_{es}/τ_P	C_e/C_P	τ_1	τ_2	P_S (10^4 W cm $^{-3}$)
A	1	0.3	0.12	0.086	2.5	16.4	0.083	7.1	3.8
	2	0.1	0.12	0.055	0.85	16.4	0.054	3.65	7.66
Sn	B	0.51	0.54	0.26	0.95	1.64	0.22	2.04	7.8

same initial temperature T_S^* has been chosen to compare the two systems. The corresponding initial powers P_S are of course different, the larger being for the B system with $P_S = 10.3 \times 10^4$ W cm $^{-3}$ [$P_S = 3.8 \times 10^4$ W cm $^{-3}$ for the A system (Table II)].

The variations $\tilde{P}_N = P_N(t) - P_{NS}$ of $P_N(t)$ are larger for the B system than for the A system. This can partly be explained by the large ratio of transverse-to-longitudinal phonon heat capacities $C_{PT}/C_{PL} \approx 10$ in Sn according to the large transverse phonon density of states $n_t(\omega)$ [$n_\alpha(\omega) \propto \omega^2 u_\alpha^{-3}$]. However, the phonon relaxation times τ_P are different in the two systems considered ($\tau_{PL} \sim 0.12$ nsec and $\tau_{PT} \sim 0.53$ nsec) and the larger τ_{PT} for transverse phonons acts directly towards a smaller $P_N(t)$, whereas through the departure of the phonons from equilibrium with the quasiparticles, it acts towards a larger $P_N(t)$. The departure of the phonon distribution from a Planck distribution at T^* , which would be the phonon distribution in a quasiparticle-phonon system in equilibrium at T^* , is represented in Fig. 4 as a function of $\hbar\omega/\Delta$ through the quantity $\omega^3[\bar{N}(T^*) - N_\alpha]$ for the A system [curve (a)] and the B system [curve (b)] for a phonon escape time $\tau_{es} = 0.3$

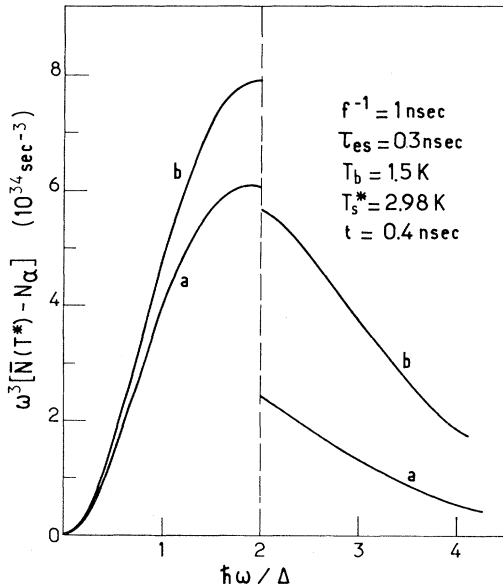


FIG. 4. Spectrum of the quantity $\omega^3[\bar{N}(T^*) - N_\alpha]$ which shows the departure of the phonon distribution from equilibrium at T^* with the quasiparticles. Curves (a) [(b)] are for the A (B) systems with longitudinal (transverse) phonons only, at time $t = 0.4$ nsec.

nsec at a time t which corresponds approximatively to the occurrence of a maximum for $P_N(t)$ in both cases (A and B) (as it is seen in Fig. 3).

In Fig. 5 is shown the energy density transferred from the quasiparticles to the phonons,

$$\omega n_\alpha(\omega)[\bar{N}(T^*) - N_\alpha] \propto u_\alpha^{-3} \omega^3 [\bar{N}(T^*) - N_\alpha],$$

in the same conditions as above, for the A system [$u_\alpha = 3.32 \times 10^3$ m s $^{-1}$; curve (a)] and for the B system [$u_\alpha = 1.95 \times 10^3$ m s $^{-1}$; curve (b)]. A comparison between the distance of curves (a) and (b) in Fig. 5 and in Fig. 4 clearly shows the influence of the phonon specific heat in the determination of $P_N(t)$ through the different state densities $n_\alpha(\omega) \propto \omega^2 u_\alpha^{-3}$.

Though the time behavior of the power $P_N(t)$ is not simple to analyze, particular attention will be paid to it, since this term is representative of the ability of the phonons to receive or give heat, and its phase shift gives information of the time response of the phonons to the excitation through the quasiparticles.

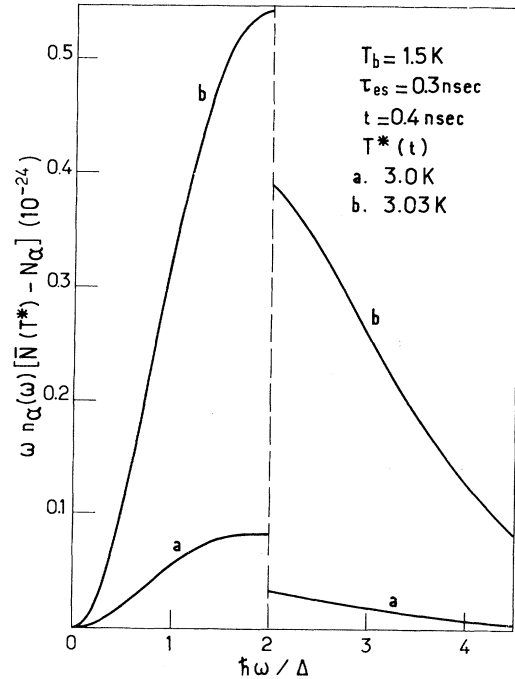


FIG. 5. Spectrum of the energy density $\omega n_\alpha(\omega)[\bar{N}(T^*) - N_\alpha]$ transferred from the quasiparticles to the phonons at time $t = 0.4$ nsec. Curves (a) [(b)] are for the A (B) systems with longitudinal (transverse) phonons only.

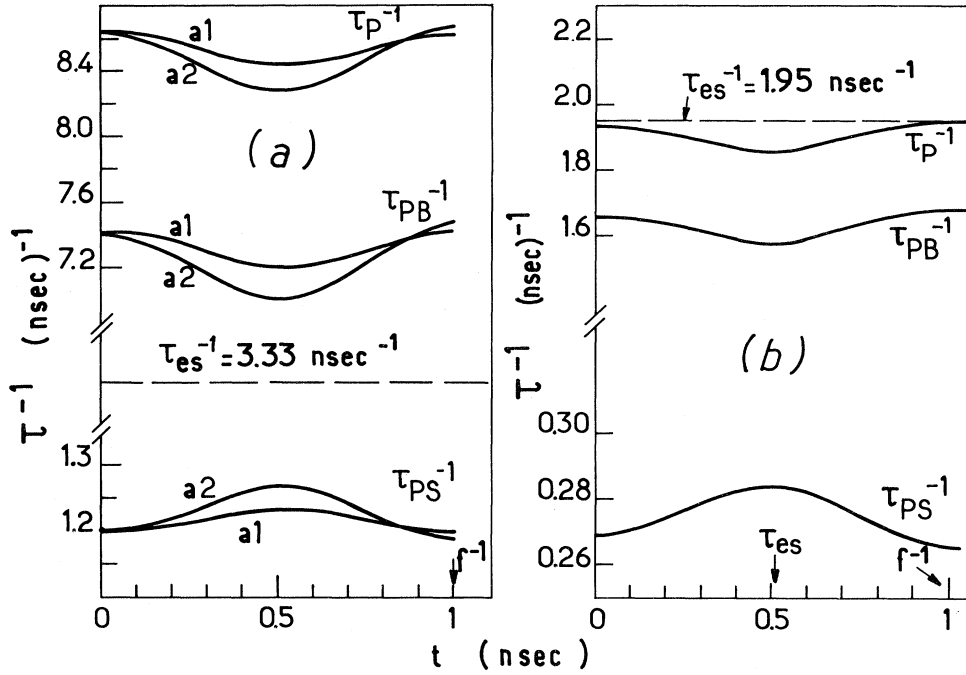


FIG. 6. Phonon relaxation rates vs time: pair-breaking relaxation rate τ_{PB}^{-1} , scattering relaxation rate τ_{PS}^{-1} for the A (B) systems [left- (right) hand side of the figure]. Curves (a1) and (a2) correspond to a phonon escape rate $\tau_{es}^{-1} = 3.33 \text{ nsec}^{-1}$ and $\tau_{es}^{-1} = 10 \text{ nsec}^{-1}$, respectively. The initial quasiparticle temperature $T_s^* = 2.98 \text{ K}$.

B. Modulation depths and phase shifts as functions of the excitation frequency

Until now we have considered a single frequency $f = 1 \text{ GHz}$. Let us now study the quantities T^* , φ_e , P_N , and φ_N as functions of the modulation frequency. In order to reduce the number of parameters in the problem, we again consider the A and B systems as above. In the A system, we consider two cases (see Table II): (1) a case with a phonon escape time $\tau_{es} = 0.3 \text{ nsec}$ and hence a situation such that

$$\tau_{PB}(2\Delta) < \tau_{es} < \tau_{PS}(2\Delta)$$

as is seen in the left part of Fig. 6 where the phonon relaxation rates $\tau_{PB}^{-1}(2\Delta)$, $\tau_{PS}^{-1}(2\Delta)$, and

$$\tau_P^{-1}(2\Delta) = \tau_{PS}^{-1}(2\Delta) + \tau_{PB}^{-1}(2\Delta)$$

are shown as functions of time t for a frequency $f = 1 \text{ GHz}$ [curves (a1)], and (2) a phonon escape time $\tau_{es} = 0.1 \text{ nsec}$ such that

$$\tau_{es} < \tau_{PB}(2\Delta) < \tau_{PS}(2\Delta)$$

and $\tau_{es} < \tau_P(2\Delta)$. As we have chosen the same initial temperature $T_s^* = 2.98 \text{ K}$, the mean value of each phonon relaxation time, $\tau_{PS}(2\Delta)$ and $\tau_{PB}(2\Delta)$, does not differ much in the two cases [curves (a1) and (a2) in Fig. 6]. The ratio of the quasiparticle to the phonon specific heat is rather large in the A system: $C_e/C_{PL} \approx 16$ (Table II). In these conditions, the quasiparticle temperature-modulation depth is shown in Fig. 7 [curves (a1) and (a2)], as a function of the excitation frequency. As expected it is shown

to decrease with the frequency f , but it must be noted that it remains rather small and far from the maximum modulation depth ($[t^*/(T_s^* - T_b)_{\max}] \rightarrow 1$ for $f \rightarrow 0$) even for the smallest frequencies considered. Indeed, the latter cor-

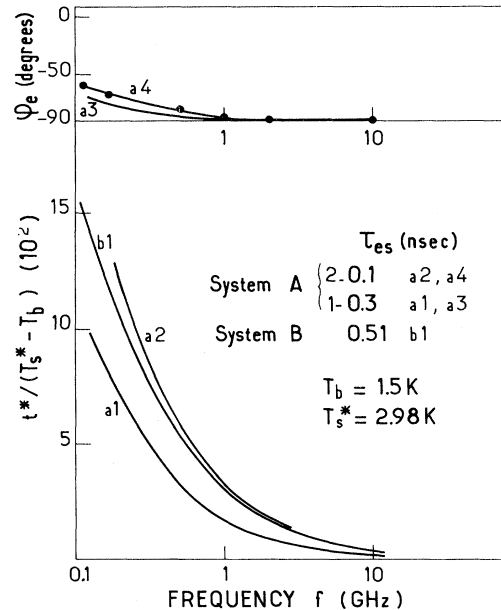


FIG. 7. Quasiparticle temperature modulation depth $t^*/(T_s^* - T_b)$ and phase shift φ_e vs drive frequency f . The filled circles on curve (a4) are the phase shifts φ_e obtained for the B system.

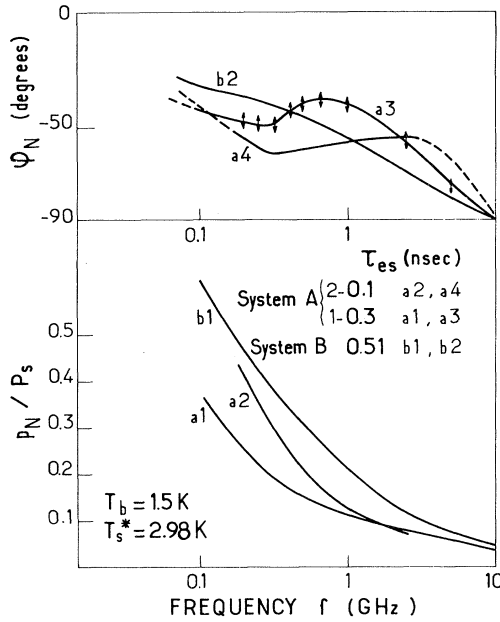


FIG. 8. Modulation depth p_N/P_S and phase shift φ_N of the power transferred from the quasiparticles to the phonons vs drive frequency f .

respond to periods $f^{-1} = 5$ and 7 nsec far larger than the mean intrinsic times of the system (see Fig. 6) which are of the order of 0.15 and 0.8 nsec, respectively, for the pair-breaking relaxation time and for the scattering relaxation time, at the temperatures considered. However, as expected, the quasiparticle temperature modulation depth is larger in case 2 [$\tau_{es} = 0.1$ nsec; curve (a2)] than in case 1 [$\tau_{es} = 0.3$ nsec; curve (a1)].

For the frequencies considered ($0.1 < f < 10$ GHz) the T^* phase shift φ_e (Fig. 7) between the quasiparticle temperature T^* and the incident power P_i for $\tau_{es} = 0.3$ nsec, remains close to $-\pi/2$ ($1 < f < 10$ GHz), the departure from this value increasing when the frequency f decreases from 1 to 0.1 GHz [curve (a3) in Fig. 7]. This is expected from the rather small variations of $P_N(t)$ as it can be seen in Fig. 8 and from Eq. (2).

For large frequencies, the quasiparticle temperature T^* is rigorously in quadrature with the incident power, since in these conditions $P_N(t)$ itself is in phase with T^* as it will be seen below. The phase shift φ_e obtained for $\tau_{es} = 0.1$ nsec (case 2) shows more important deviations from $-\pi/2$ [curve (a4) in Fig. 7]. This is expected from the larger variation of $P_N(t)$ and from Eq. (2). Therefore, as the phonon trapping in the film decreases, the importance on the quasiparticle-system behavior of the energy transfer from the quasiparticles to the phonon system increases.³⁵

The phase shift φ_N and the $P_N(t)$ modulation depth p_N/P_S are shown in Fig. 8 as functions of the excitation frequency f . The modulation depth p_N/P_S (lower curves) decreases monotonically with f in the frequency range considered. It is larger for the phonon escape time $\tau_{es} = 0.1$ nsec [curve (a2)] than for $\tau_{es} = 0.3$ nsec [curve (a1)]. This is well understood since a smaller phonon escape time τ_{es} means an easier transmission of the phonon

energy to the substrate; when $\tau_{es} = 0.3$ nsec $> \tau_P$ (case 1), the quasiparticle and phonon systems are closer to an equilibrium system in which there is no more transfer of energy from quasiparticles to phonons. For very small excitation frequencies, p_N/P_S must tend towards 1 since at each time, the system is then in a quasistationary state.

The phase shift φ_N exhibits a nonmonotonic behavior between the two extreme values $-\pi/2$ ($f \rightarrow \infty$) and zero ($f = 0$) as is seen in Fig. 8 [curves (a3) and (a4)]. For large frequencies, the phonon distribution remains nearly constant since $\partial N/\partial t \simeq 0$ and P_N is entirely determined by the quasiparticle temperature T^* ; it is then in quadrature with the incident power as T^* itself. From these results, it seems that the phase-shift behavior is more striking than the amplitude behavior.

The same kind of behavior is observed for $\tau_{es} = 0.1$ nsec. However, the positions of the maximum and minimum are different. From Figs. 7 and 8 together, we can deduce that the response of the film is better in case 2 than in case 1.

Let us now consider the B system (transverse phonons only) with a single phonon escape time $\tau_{es} = 0.51$ nsec (Table II). The coupling of the transverse phonons with the quasiparticles leads to larger quasiparticle-phonon relaxation times as can be seen in Fig. 6. The phonon escape time $\tau_{es} = 0.51$ nsec is such that $\tau_{es} \sim \tau_P(2\Delta)$, $\tau_{es} < \tau_{PB}(2\Delta) < \tau_{PS}(2\Delta)$ (right part in Fig. 6). We thus have an intermediate situation between the two cases just considered. As we mentioned before, the ratio of the quasiparticle and phonon specific heat in this case is $C_e/C_{PT} \simeq 1.6$ (Table II) and therefore smaller than above (case 1 and case 2).

As can be seen in Fig. 8 [curve (b1)], the modulation depth p_N/P_S of the power transmitted to the phonons is larger than in case 1 and case 2 [curves (a)] though the ratio τ_{es}/τ_P (Table II) is larger than for case 2 [curve (a2)]. However, it must be borne in mind that the phonon specific heat is here an order of magnitude larger than for case 2 ($C_{PT}/C_{PL} \simeq 10$), which explains the larger p_N/P_S obtained. The phase shift φ_N [curve (b2) in Fig. 8] is either larger or smaller than in case 1 and case 2 above (A system; longitudinal phonons only), according to the frequency. But, the most striking fact is the quite different behavior of φ_N , which is here a monotonic function of the frequency in the range studied.

The quasiparticle temperature modulation depth is shown in Fig. 7 [curve (b1)], as a function of the frequency. Its behavior is quite similar to the behavior obtained in case 1 and case 2 [curves (a); A system]. However, the calculated values are far nearer to curve (a2) ($\tau_{es}/\tau_P < 1$; Table II) than to curve (a1) ($\tau_{es}/\tau_P > 1$). This again results from the large phonon specific heat. The quasiparticle phase shift φ_e (filled circles on the upper curve in Fig. 7) is quite similar to the phase shift φ_e in the A-2 case above in the range of frequencies considered.

In the above analysis, we have seen that a large value of the quasiparticle specific heat C_e compared to the phonon specific heat C_P ($C_e/C_P > 1$) leads to a small response of the system, essentially due to the small amount of energy transferred to the phonon system. This was particularly clear in the 1-GHz modulation study and was confirmed by the frequency analysis [Fig. 8; curves (a1) and (b1)]; indeed, the quasiparticle temperature phase shift φ_e

remains close to $-\pi/2$, and therefore nearly independent on the energy transferred to the phonons, for a large range of frequencies. As the ratio τ_{es}/τ_P decreases, there is a larger possibility of energy transfer to the phonons [curves (a1) and (a2) in Fig. 7]. In the Sn film study just above, the phonon response is seen to be essentially determined by the ratio C_e/C_P whereas the quasiparticle response is very sensitive to the ratio τ_{es}/τ_P [comparison of curves (a2) and (b1) in Figs. 7 and 8].

Moreover, the nonmonotonic behavior of φ_N observed in certain conditions makes us guess that there are characteristic frequencies in the system that are different from the frequencies τ_{es}^{-1} and τ_P^{-1} . It also shows that previous qualitative predictions about the response of the superconducting film are far too simple.²¹

It would be interesting to determine exactly the characteristic times of the system. Moreover, in considering other materials such as Pb with $C_e/C_P < 1$, we could get more information. Unfortunately, the calculation would be very long even with the quasiparticle T^* Fermi-Dirac distribution approximation, particularly for materials which intrinsic times are short compared to τ_{es} . So, let us introduce a simple analytic model that can be used, as is shown below, in the study of other materials behavior, which is beyond the scope of this paper and will be done elsewhere.³⁶

V. COMPARISON

In this model, the phonon system is characterized by a single temperature $T_P(t)$ and a single constant relaxation time τ_P . The equations equivalent to Eqs. (2) and (8) above are then

$$C_e(dT^*/dt) = P_i - C_e(T^* - T_P)/\tau_e \quad (13)$$

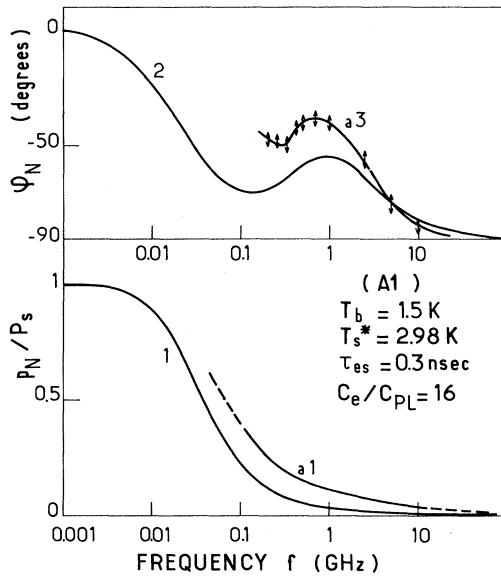


FIG. 9. Modulation depth p_N/P_S and phase shift φ_N of the power transferred from the quasiparticles to the phonons vs drive frequency f for the analytic model [curves (1) and (2)] and for our numerical calculations [curves (a1) and (a3)] in the A system (longitudinal phonons only).

and

$$C_P(dT_P/dt) = C_P(T^* - T_P)/\tau_P - C_P(T_P - T_b)/\tau_{es} \quad (14)$$

From detailed balance, we have $C_e/\tau_e = C_P/\tau_P$, τ_e being the quasiparticle relaxation time by phonon scattering. In these conditions, the amplitudes $t^* = T_{\max}^* - T_S^*$ and $p_N = P_{N\max} - P_S$ can be expressed in the complex form by

$$t_c^* = P_S \frac{\tau_P + \tau_{es}}{C_P} \frac{1 + j\omega_i \tau_\varphi}{(1 + j\omega_i \tau_1)(1 + j\omega_i \tau_2)} \quad (15)$$

and

$$p_{NC} = P_S \frac{1 + j\omega_i \tau_{es}}{(1 + j\omega_i \tau_1)(1 + j\omega_i \tau_2)}, \quad (16)$$

with

$$\tau_{1,2}^{-1} \equiv \tau_{+,-}^{-1} = \frac{1}{2\tau} \left[1 \pm \left[1 - 4 \frac{\tau^2}{\tau_{es}\tau_P} \frac{C_P}{C_e} \right]^{1/2} \right], \quad (17)$$

$$\tau^{-1} = \tau_{es}^{-1} + \tau_P^{-1}(1 + C_P/C_e), \quad (18)$$

and

$$\tau_\varphi^{-1} = \tau_P^{-1} + \tau_{es}^{-1}. \quad (19)$$

From these equations, it is seen that, besides the intrinsic times, there exists characteristic times in the system, τ_1 and τ_2 , which are functions of the former times.

The modulation depths and phase shifts corresponding to Eqs. (15) and (16) are shown in Figs. 9–11 along with our results for the A system in case 1 (Figs. 9 and 11) and for the B system (Fig. 10). The type of behavior obtained with this model is quite similar to the behavior obtained before with the full numerical calculations, except for the quantitative values which differ slightly. Therefore, from

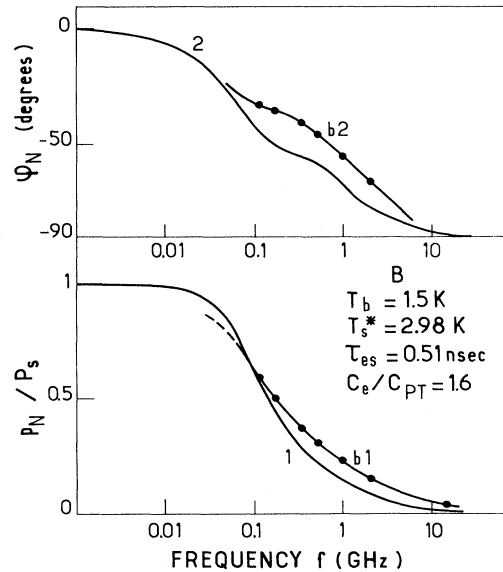


FIG. 10. Modulation depth p_N/P_S and phase shift φ_N of the power transferred from the quasiparticles to the phonons vs drive frequency f , for the analytic model [curves (1) and (2)] and for our numerical calculations [curves (b1) and (b2)] in the B system (transverse phonons only).

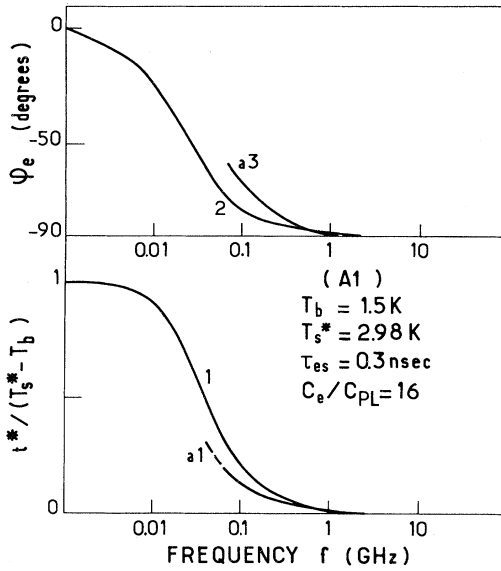


FIG. 11. Quasiparticle temperature modulation depth $t^*(T_s^* - T_b)$ and phase shift φ_e vs drive frequency f for the analytic model [curves (1) and (2)] and for our numerical calculations [curves (a1) and (a3)] in the A system (longitudinal phonons only).

Eqs. (15) and (16), we can guess that the behavior of p_N and t^* are determined by the relative values of the characteristic times τ_1 and τ_2 compared to τ_{es} and τ_φ , respectively. These times τ_1 and τ_2 are shown in Table II.

Let us first consider the phase shifts φ_N and φ_e . From Eq. (16) it is seen that for small frequencies such that $\omega_i\tau_{es} \ll 1$, $\omega_i\tau_{1,2} \ll 1$, the phase shift φ_N remains close to zero. When the frequency is increased, the largest of the characteristic times τ_2 will cause the phase shift φ_N to decrease towards $-\pi/2$ for frequencies ω_i such that $\omega_i\tau_2 \approx 1$ as long as $\omega_i\tau_{1,es} \ll 1$. Then according to the relative values of τ_{es} and τ_1 , it may behave differently, either exhibiting a nonmonotonic behavior when $\tau_1 \ll \tau_{es}$ [A system, case 1; curves (a3) and (2) in Fig. 9] or a monotonic behavior for $\tau_1 \approx \tau_{es}$. This type of behavior can also be obtained for $\tau_1 < \tau_{es} < \tau_2$, if these times are not too different from each other; this is the case of the B system as can be seen in Table II and Fig. 10 [curves (b2) and (2)]. Moreover, the frequency for which the phase shift will nearly reach the zero value (which characterizes the response of the film phonons) is the larger as the three characteristic times are small. This explains the rather fast response obtained for the B system. The phase shift φ_e of the quasiparticle temperature exhibits a monotonic behavior [see Figs. 11, curves (a3) and (2), and Fig. 7] since in all the cases considered above, the time τ_φ in the expression of φ_e is close enough to τ_1 .

As can be seen from Eqs. (15) and (16), the amplitude of the response for the power $P_N(t)$ as well as for the quasiparticle temperature T^* are governed by the same characteristic times τ_1 and τ_2 . With the values of Table II, it can be shown from Eqs. (15) and (16) that the amplitudes t^* and p_N are decreasing functions of the frequency for all the systems considered above. This agrees with the results of our computer analysis over the range of frequency

0.1–10 GHz which can be seen in Figs. 9–11 for the A system (case 1) and for the B system. From the expressions of the characteristic times τ_1 and τ_2 in Eqs. (17) and (18) the importance of the specific-heat ratio C_e/C_p is clearly seen. Therefore, it would be interesting to study materials with different values of the ratio C_e/C_p . A detailed study of the response of different materials in the scope of this simple analytic model will be presented in another paper.³⁶

VI. DISCUSSION AND CONCLUSION

The response of the system of quasiparticles and phonons to a periodic laser irradiation, is seen to be slow in a superconducting Sn film, which behavior is expected to be determined essentially by the transverse phonons. The quasiparticle temperature modulation depth remains small for the smallest frequency considered.

From the frequency study of the quasiparticle temperature T^* and of the power P_N received by the phonons through the quasiparticles, it is clearly seen that the phonon escape time does not determine the film response so directly as it has been suggested in previous work.^{18,21} In fact, it is the largest of the characteristic times $\tau_{1,2}$ and τ_φ which first perturbs the response of the quasiparticle system and therefore the modulation of the superconducting film properties through the gap modulation. The times τ_1 and τ_2 depend both on the phonon escape time τ_{es} and on the phonon relaxation time τ_p . In order to analyze the part played by these different times, we have studied the frequency dependence of the amplitude and of the phase of the power $P_N(t)$ transferred to the phonon system by the quasiparticles. This frequency dependence exhibits a behavior which is characteristic of the response of both quasiparticle and phonon populations. The times τ_1 and τ_2 also depend on the ratio C_e/C_p of the quasiparticle and the phonon specific heat, the largest characteristic time τ_2 decreasing as C_e/C_p decreases in the cases studied.

In the case of Sn just considered, the response of the quasiparticle system is restricted by the energy exchanges between the quasiparticles and the phonons. This was clearly seen when we considered the longitudinal phonons only, with the largest phonon escape time $\tau_{es} = 0.3$ nsec; in this case, most of the energy remains in the quasiparticle system and the film response is particularly slow and small. However, in Sn, the transverse phonons yield a faster response than the longitudinal ones, according to their smaller ratio C_e/C_p . For very small C_e/C_p (which is the case for lead where $C_e/C_p \approx 0.07$), the energy provided by the laser irradiation is expected to be essentially transferred to the phonon system. (Whereas it is trapped in the quasiparticle system for $C_e/C_p > 1$.) In these conditions, the intrinsic times of the phonon system, and particularly the phonon escape time, become determinant in the system response.

Let us note that in our calculations, the laser irradiation is described by a quasiparticle injection term. This is quite valid for the Sn film considered, but in lead, according to the small ratio of the quasiparticle and phonon relaxation times ($\tau_e/\tau_p \ll 1$), a phonon injection term would probably be necessary.

We have shown that the results of a simplified model for the phonons yields qualitative results which agree with our full numerical calculations. This allows us to deter-

mine analytically the excitation frequency for which a maximum amplitude modulation of the quasiparticle temperature and of the gap is obtained, as a function of the characteristic times of the system. In Sn this frequency

$\omega_M = \tau_2^{-1}$ is smaller than the frequency τ_{es}^{-1} . Information concerning the qualitative time behavior of any superconducting film could therefore be deduced from this model.³⁶

- ¹L. R. Testardi, Phys. Rev. B **4**, 2189 (1971).
²W. H. Parker and W. D. Williams, Phys. Rev. Lett. **28**, 924 (1972).
³P. Hu, R. C. Dynes, and V. Narayanamurti, Phys. Rev. B **10**, 2786 (1974).
⁴B. Lambert, D. Salin, and J. Joffrin, J. Phys. (Paris) Lett. **38**, L 343 (1977).
⁵F. Jaworski and W. H. Parker, Phys. Rev. B **20**, 945 (1979).
⁶O. P. Balkashin, A. V. Khotkevich, and I. K. Yanson, Fiz. Nizk. Temp. **5**, 28 (1979) [Sov. J. Low Temp. Phys. **5**, 12 (1979)].
⁷A. D. Smith, W. J. Skocpol, and M. Tinkham, Phys. Rev. B **21**, 3879 (1980).
⁸G. A. Sai-Halasz, C. C. Chi, A. Denenstein, and D. N. Langenberg, Phys. Rev. Lett. **33**, 215 (1974).
⁹A. I. Golovashkin, K. V. Mitsen, and G. P. Motulevich, Zh. Eksp. Teor. Fiz. **68**, 1408 (1975) [Sov. Phys.—JETP **41**, 701 (1975)]; A. I. Golovashkin, O. M. Ivanenko, K. V. Mitsen, G. P. Motulevich, and A. A. Shubin, Zh. Eksp. Teor. Fiz. **75**, 1517 (1979) [Sov. Phys.—JETP **48**, 766 (1978)].
¹⁰R. Buisson, R. Chicault, F. Madeore, and R. Romestain, J. Phys. (Paris) Lett. **39**, L130 (1978); J. Phys. (Paris) Suppl. **39**, C6-509 (1978).
¹¹L. N. Smith, J. Low Temp. Phys. **38**, 553 (1980).
¹²W. H. Parker, Solid State Commun. **15**, 1003 (1974).
¹³P. Hu, R. C. Dynes, V. Narayanamurti, and W. F. Brinkman, Phys. Rev. Lett. **38**, 361 (1977); V. Narayanamurti, R. C. Dynes, P. Hu, H. Smith, and W. F. Brinkman, Phys. Rev. B **18**, 6041 (1978).
¹⁴F. Jaworski, W. H. Parker, and S. B. Kaplan, Phys. Rev. B **14**, 4209 (1976).
¹⁵I. Schuller and K. E. Gray, Phys. Rev. B **12**, 2629 (1975); Phys. Rev. Lett. **36**, 429 (1976).
¹⁶P. W. Epperlein, K. Lassmann, and W. E. Eisenmenger, Z. Phys. B **31**, 377 (1978).
¹⁷C. C. Chi, M. M. T. Loy, and D. C. Cronmeyer, Phys. Rev. B **23**, 124 (1981).
¹⁸C. C. Chi, M. M. T. Loy, D. C. Cronmeyer, and M. L. Thewalt, IEEE Trans. Mag. **17**, 88 (1981).
¹⁹R. Sobolewski, private communication.
²⁰H. Seifert, J. Low Temp. Phys. **37**, 597 (1979).
²¹C. Vanneste, A. Gilabert, P. Sibillot, and D. B. Ostrowsky, J. Low Temp. Phys. **45**, 517 (1981).
²²J. J. Chang, W. Y. Lai, and D. J. Scalapino, Phys. Rev. B **20**, 2379 (1979).
²³N. Perrin, J. Phys. **40**, 1089 (1979); J. Phys. **41**, 615 (1980).
²⁴A. Rothwarf and B. N. Taylor, Phys. Rev. Lett. **19**, 27 (1967).
²⁵S. B. Kaplan, C. C. Chi, D. N. Langenberg, J. J. Chang, S. Jafarey, and D. J. Scalapino, Phys. Rev. B **14**, 4854 (1976).
²⁶J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).
²⁷R. C. Dynes, V. Narayanamurti, and M. Chin, Phys. Rev. Lett. **26**, 181 (1971).
²⁸A. R. Long and C. J. Adkins, Philos. Mag. **27**, 865 (1973).
²⁹W. H. Parker, Phys. Rev. B **12**, 3667 (1975).
³⁰G. Rickayson, *Theory of Superconductivity* (Wiley, New York, 1964), p. 205; M. Tinkham, *Documents on Modern Physics—Superconductivity* (Gordon and Breach, New York, 1965), p. 43.
³¹N. Perrin, Solid State Commun. **17**, 131 (1974).
³²B. Pannetier and J. P. Maneval, Phys. Rev. B **23**, 85 (1981).
³³The small decay observed is due to the initial conditions imposed.
³⁴For phonons of energy $\hbar\omega > 2\Delta$, both pair-breaking and quasiparticle phonon scattering determine the phonon relaxation time τ_P (Eq. 5), whereas for $\hbar\omega < 2\Delta$, $\tau_P \equiv \tau_{PS}$. Since $\tau_{PB} < \tau_{PS}$ and they both decrease with the phonon frequency ω (Ref. 23), the shortest values of τ_P occur for phonon energies $\hbar\omega > 2\Delta$. However, as the frequency ω increases, the departure from an equilibrium between the phonon system and the quasiparticle system decreases (as can be seen in Fig. 4), and we can guess that the relevant time in the determination of $P_N(t)$ is the relaxation time τ_P in the neighborhood of $\hbar\omega = 2\Delta$, which will be referred to.
³⁵U. Eckern and G. Schön, J. Low Temp. Phys. **32**, 821 (1978).
³⁶A detailed analysis of this model will be presented elsewhere.