

## Transient electrodynamic response of thin-film superconductors to laser radiation

N. E. Glass and D. Rogovin

*Rockwell International Science Center, 1049 Camino Dos Rios, Thousand Oaks, California 91360*

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We examine a theory of the transient electrodynamics of thin-film superconductors irradiated by both cw- and pulsed-laser light. The theory employs the Rothwarf-Taylor rate equations in conjunction with a time-dependent version of Parker's  $T^*$  model of nonequilibrium superconductivity, to determine the microwave and far-infrared electrodynamics. This formulation is sufficiently general that it can also be utilized for problems in which either static or dynamic gratings are written on the superconductor by two laser beams. We apply the theory to studies of time-dependent microwave transmission through thin superconducting films and microwave propagation on superconducting waveguides. A perturbative formulation of the electrodynamics, to first order in the quasiparticle response, is also presented.

### I. INTRODUCTION

The study of superconductors whose quasiparticle (QP) and phonon densities have been driven out of equilibrium by external fields or currents is of considerable interest in low-temperature and device physics. This interest is motivated by the possible appearance of novel physical phenomena as well as by device applications. An extensive review of the physics of nonequilibrium superconductivity can be found in Refs. 1 and 2.

A particularly interesting application is the optical control of microwaves by use of lasers. Here, visible laser light splits pairs creating an excess of QP's, a process which depresses the gap and alters the QP distribution. These effects, in turn, change the superconductor's conductivity for frequencies below twice the gap frequency. Lasers might thus be employed to control the propagation characteristics of microwaves on a strip line. Pulsed lasers could modulate the amplitude or phase of the microwave radiation. Furthermore, a superconductor irradiated by two degenerate laser beams might form a static electromagnetic grating, whose spatial period is set by the incident laser wavelengths and orientations. Such a grating could be utilized to guide microwave or far-infrared radiation on a thin film. If the laser beams are nondegenerate, the electromagnetic gratings will oscillate at the difference frequency, so that the superconductor, again, is not in the steady state. One is thus motivated to investigate problems in which the driving currents vary in time and/or space—not only for these potential device applications, but also because the behavior of a many-body system under such nonequilibrium situations is of interest to statistical mechanics. In particular, spatially nonuniform nonequilibrium superconductivity has received much attention in recent years in the attempt to understand diffusive instabilities.

In this paper we employ a theory of nonequilibrium time-dependent superconductivity which is appropriate for describing thin-film superconductors irradiated by either cw- or pulsed-laser light emitted by one or more

sources. Our principal goal is to determine the transient-microwave conductivity for such situations, especially for cases when the thin film is driven far from equilibrium by the laser pulse. Once the conductivity is obtained it is straightforward to determine the transient-microwave transmission characteristics of a thin-film superconductor or the propagation characteristics of a superconducting strip line.

It is difficult to treat, by first principles, superconducting systems that are driven far from equilibrium by fields whose amplitudes vary in time and/or space. A variety of *phenomenological* schemes, however, are presently available for treating superconductors far from equilibrium. One category of such schemes is based on the Rothwarf-Taylor (RT) rate equations, which treat the kinetics of the QP and phonon densities.<sup>3</sup> We utilize the RT equations in a form including diffusion, which occurs whenever the superconductor is driven by inhomogeneous fields, as when an electromagnetic grating is written on a thin film. Since we are interested in the microwave response, i.e., wavelengths greater than a millimeter, we shall not be concerned here with the *intrinsic* diffusion-driven instabilities,<sup>4</sup> which manifest themselves on spatial scales on the order of 100  $\mu\text{m}$  or less. The problem is to solve simultaneous partial differential equations for the QP and phonon densities as functions of time  $t$  and position  $\mathbf{r}$ :  $n_{\text{QP}}(\mathbf{r}, t)$  and  $n_{\text{ph}}(\mathbf{r}, t)$ , respectively. We confine ourselves to situations in which the phonon response time is much shorter than that of the QP, so that the phonons will adiabatically follow the QP. For external fields that vary sufficiently slowly in time and space, the phonon coordinate can be adiabatically eliminated. This approximation leaves us with only one differential equation to solve, thus considerably simplifying the problem. The simplification allows us to develop an analytic solution for a general space- and time-varying drive current when the system is not far from equilibrium, and an analytic solution for a spatially uniform but time-pulsed drive when the system is far from equilibrium.

Evaluating the nonequilibrium electrodynamic re-

sponse of the superconductor requires knowing the QP energy distribution and the gap energy. For situations of interest to us—far from equilibrium where the QP response may be highly nonlinear in the drive fields or where the drive fields are spatially nonuniform—it is impractical to use an approach based on the microscopic kinetic (Boltzmann) equations.<sup>5,6</sup> As we are interested principally in optical laser generation of the QP's (which relax rapidly to a thermal distribution), a time-dependent version of Parker's effective-temperature ( $T^*$ ) model<sup>7</sup> is employed.

The nonequilibrium gap and QP energy distribution are then used in the Mattis-Bardeen expression for the complex dynamic conductivity,<sup>8</sup> in order to determine the electrodynamic response, at frequencies near the gap. We present numerical results for the time-dependent microwave conductivity, thin-film transmittance, and waveguide propagation, in response to an optical laser pulse.

We find that over a fairly wide range of parameters the change in the electrodynamic response due to the applied fields is a linear function of the fractional change in the energy gap, even though the latter quantity may be a nonlinear function of the driving fields. This feature of superconducting electrodynamics allows us to develop a convenient perturbation theory for the change in the electrodynamic response, to first order in the change in QP density.

The paper is organized as follows: Section II is devoted to the kinetics of the QP and phonon densities. In Sec. II A we present the RT rate equations and discuss the adiabatic elimination of the phonon variable. In Sec. II B we offer solutions of these equations for some specific time- and position-dependent field amplitudes. In Sec. III we discuss  $T^*$  and the nonequilibrium energy gap obtained from the QP density. Section IV is devoted to electrodynamics. In Sec. IV A we outline the method for determining the electrodynamic response functions from the Mattis-Bardeen equations and the  $T^*$  kinetics. We present results for the changes in the microwave transmission and waveguiding characteristics under intense optical irradiation—both steady state and pulsed. In Sec. IV B we present a theory which determines the electrodynamic response of a superconductor, to first order in the fractional change in energy gap, and thus to first order in the change in QP density. We discuss the limits of validity of this linear relationship. Mathematical details of this first-order theory are presented in the Appendix. We conclude in Sec. V.

## II. KINETICS

The physical situation to which this model applies is as follows: A superconducting thin film lies on a substrate, such as sapphire, which also serves as a thermal reservoir. The thin film is subjected to QP and phonon currents which drive the system out of thermodynamic equilibrium. These currents arise from external drive fields and can vary in time and space, as does the external-field intensity. In the special case in which the

external drive is a laser field, it is possible to consider the QP current alone, and relate it to the laser intensity.

### A. The rate equations: Adiabatic elimination of the phonons

Our starting point is a generalization of the RT equations,<sup>3</sup> to include the effects of QP and phonon diffusion,<sup>2</sup> which arises from spatially inhomogeneous drive currents (e.g., laser induced). The RT equations are essentially generation-recombination equations. Hence, diffusion enters into the QP (or phonon) equation via the divergence of a QP (or phonon) diffusion current  $J_{\text{QP}}(\mathbf{r}, t)$  [or  $J_{\text{ph}}(\mathbf{r}, t)$ ], where

$$J_{\text{QP}}(\mathbf{r}, t) = D_{\text{QP}} \nabla n_{\text{QP}}(\mathbf{r}, t), \quad (2.1a)$$

$$J_{\text{ph}}(\mathbf{r}, t) = D_{\text{ph}} \nabla n_{\text{ph}}(\mathbf{r}, t). \quad (2.1b)$$

In these equations,  $D_{\text{QP}}$  and  $D_{\text{ph}}$  are phenomenological diffusion coefficients for the QP and phonons. Accordingly, the RT equations become

$$\left[ \frac{\partial}{\partial t} - D_{\text{QP}} \nabla^2 \right] n_{\text{QP}} = I_{\text{QP}} - 2Rn_{\text{QP}}^2 + \frac{2}{\tau_B} n_{\text{ph}}, \quad (2.2a)$$

$$\left[ \frac{\partial}{\partial t} - D_{\text{ph}} \nabla^2 \right] n_{\text{ph}} = I_{\text{ph}} + Rn_{\text{QP}}^2 - \frac{1}{\tau_B} n_{\text{ph}} - \left[ \frac{n_{\text{ph}} - n_{\text{ph}}^0}{\tau_{\text{es}}} \right]. \quad (2.2b)$$

In Eqs. (2.2),  $I_{\text{QP}}$  and  $I_{\text{ph}}$  are the drive currents per unit volume due to the external fields;  $R$  is the recombination coefficient;  $\tau_B$  is the time for a phonon to break a pair;  $\tau_{\text{es}}$  is the time for a phonon to escape to the thermal reservoir; and  $n_{\text{ph}}^0$  is the equilibrium phonon density associated with the thermal reservoir. The quantity  $(2Rn_{\text{QP}})^{-1}$  is regarded as a quasiparticle-recombination time.

Next we estimate the space and time scales over which the QP and phonon densities vary. This can be accomplished by scaling the densities as well as the space and time coordinates to dimensionless quantities. With  $n_{\text{QP}}^{\text{SS}}$  and  $n_{\text{QP}}^0$  defined as the QP density in steady-state (SS) and in equilibrium, respectively, we introduce  $u$  as

$$u = n_{\text{QP}} / n_{\text{QP}}^{\text{SS}} \quad (2.3a)$$

or

$$u = n_{\text{QP}} / n_{\text{QP}}^0, \quad (2.3b)$$

depending on whether the final state is, or is not, the equilibrium state. For the phonons, we normalize to the equilibrium thermal reservoir value, as

$$v = n_{\text{ph}} / n_{\text{ph}}^0. \quad (2.4)$$

The natural time scale to use is

$$\tau_R = (2Rn_{\text{QP}}^{\text{SS}})^{-1} \quad (2.5a)$$

or

$$\tau_R^0 = (2Rn_{\text{QP}}^0)^{-1}, \quad (2.5b)$$

depending again on the final state of the system. With respect to the steady-state values, we introduce the dimensionless time as

$$s = t / \tau_R . \quad (2.6)$$

The natural length scale to use is the QP recombination length

$$L_{QP} = (D_{QP} \tau_R)^{1/2} , \quad (2.7)$$

and accordingly, we introduce the following definition:

$$\underline{\nabla} = L_{QP} \nabla . \quad (2.8)$$

When the final state is equilibrium, we replace  $\tau_R$  by  $\tau_R^0$  in Eqs. (2.6)–(2.8). Using these definitions, with the steady-state values for  $u$  and  $\tau_R$ , we find that the RT equations become

$$\left[ \frac{\partial}{\partial s} - \underline{\nabla}^2 \right] u = \frac{\tau_R I_{QP}}{n_{QP}^{SS}} - u^2 + \frac{2\tau_R n_{ph}^0}{\tau_B n_{QP}^{SS}} v , \quad (2.9a)$$

$$\left[ \frac{\tau_{ph}}{\tau_R} \frac{\partial}{\partial s} - \left[ \frac{L_{ph}}{L_{QP}} \right]^2 \underline{\nabla}^2 \right] v = \left[ \frac{I_{ph}}{n_{ph}^0} + \frac{1}{\tau_{es}} \right] \tau_{ph} + \frac{\tau_{ph} n_{QP}^{SS}}{2\tau_R n_{ph}^0} u^2 - v . \quad (2.9b)$$

Here we have introduced the phonon lifetime

$$\tau_{ph} = \frac{\tau_B \tau_{es}}{\tau_B + \tau_{es}} , \quad (2.10)$$

and length scale

$$L_{ph} = (D_{ph} \tau_{ph})^{1/2} . \quad (2.11)$$

If the phonon density is adiabatically to follow the QP density, we require that  $\tau_{ph} \ll \tau_R$  and  $L_{ph} \ll L_{QP}$  and also require that the phonon drive current  $I_{ph}$  vary in time and space on scales that are long compared to  $\tau_{ph}$  and  $L_{ph}$ .

We can estimate the time scales as follows: Conventional theory asserts that  $\tau_R \gg \tau_B$  for  $T \ll T_c$ , which implies that  $\tau_R \gg \tau_{ph}$ , since the phonon lifetime is less than the smaller of  $\tau_B$  or  $\tau_{es}$  [as confirmed by Eq. (2.10)]. Thus, even if there is weak coupling to the reservoir (which, in fact, we shall require for the  $T^*$  model, whose validity depends on the condition  $\tau_{es} \gg \tau_B$ ), it is nonetheless true that the phonon lifetime is shorter than that of the QP.

We can estimate the diffusion length scales from the approximate velocities and lifetimes of the quasiparticles and phonons, as

$$\frac{L_{ph}}{L_{QP}} \simeq \frac{v_{ph} \tau_{ph}}{v_F \tau_R} \ll 1 , \quad (2.12)$$

where the inequality follows from the previous inequality between time scales and from  $v_{ph} \ll v_F$ , where  $v_F$  is the Fermi velocity.

Thus, we anticipate that the phonon density will adiabatically follow that of the quasiparticles. Accordingly, if

the phonon current varies slowly on the scales set by  $\tau_{ph}$  and  $L_{ph}$ , then the left-hand side of Eq. (2.9b) can be set equal to zero and

$$v = \left[ \frac{I_{ph}}{n_{ph}^0} + \frac{1}{\tau_{es}} \right] \tau_{ph} + \frac{\tau_{ph}}{2\tau_R} \frac{n_{QP}^{SS}}{n_{ph}^0} u^2 . \quad (2.13)$$

Inserting Eq. (2.13) into (2.9a), we obtain, after returning to dimensional space and time coordinates,

$$\left[ \tau_R \frac{\partial}{\partial t} - L_{QP}^2 \nabla^2 \right] u = J_i \tau_R - \frac{1}{1 + \tau_{es}/\tau_B} u^2 , \quad (2.14)$$

where

$$J_i = \frac{I_{QP}}{n_{QP}^{SS}} + 2 \frac{\tau_{es}}{\tau_B + \tau_{es}} \left[ I_{ph} + \frac{n_{ph}^0}{\tau_{es}} \right] \frac{1}{n_{QP}^{SS}} . \quad (2.15)$$

One further simplification is possible. If we redefine  $u$  with respect to the equilibrium QP density [Eq. (2.3b)] instead of the steady-state value, then the above equations, (2.14) and (2.15), still hold providing that  $n_{QP}^{SS}$  is replaced by  $n_{QP}^0$  and  $\tau_R$  is replaced by  $\tau_R^0$ . Then, when the system is in equilibrium, so that  $I_{QP} = I_{ph} = 0$ , and  $u = 1$ , the resulting two equations for  $u$  and  $v$  yield the result that

$$\frac{n_{ph}^0}{\tau_B} = \frac{n_{QP}^0}{2\tau_R^0} , \quad (2.16)$$

exactly as required by detailed balance. With this result, Eq. (2.14) can be simplified to

$$\left[ \frac{\partial}{\partial t} - D_{QP} \nabla^2 \right] n_{QP} = \left[ I_{QP} + \frac{2\tau_{es}}{\tau_B + \tau_{es}} I_{ph} \right] - \frac{2R}{(1 + \tau_{es}/\tau_B)} [n_{QP}^2 - (n_{QP}^0)^2] , \quad (2.17a)$$

and Eq. (2.13) becomes

$$n_{ph} = \frac{n_{ph}^0}{(1 + \tau_{es}/\tau_B)} + I_{ph} \tau_{ph} + R \tau_{ph} n_{QP}^2 . \quad (2.17b)$$

We thus have a single differential equation to solve, Eq. (2.17a), for the QP density as a function of  $\mathbf{r}$  and  $t$ . The resulting solution  $n_{QP}(\mathbf{r}, t)$  is then to be substituted into the algebraic equation (2.17b) for the phonon density  $n_{ph}(\mathbf{r}, t)$ .

We shall be exploring a model of laser radiation in which  $I_{ph} = 0$ , and  $I_{QP}$  is proportional to the laser intensity. Note that in the absence of a phonon current, Eq. (2.17b) reduces to the nonequilibrium generalization of detailed balance

$$\frac{n_{ph}}{\tau_{ph}} = \frac{n_{QP}}{2\tau_R} , \quad (2.18)$$

provided that  $\tau_B \ll \tau_{es}$ . Equation (2.18) has useful implications regarding transient behavior, which will be discussed later.

Equations (2.17), our working equations for the QP and phonon densities, are valid (as are the RT equations) pro-

vided that the number of superconducting pairs greatly exceeds the number of QP; and this condition implies roughly that  $T/T_c \leq 0.7$ .

### B. Quasiparticle response to laser radiation

We now obtain solutions to the above rate equation, for the QP, for three interesting cases. The phonon solutions follow from Eq. (2.17b). We present our results in terms of an effective drive current,  $Q$ , which, in general, is a function of  $I_{QP}$  and  $I_{ph}$ . In Sec. II C we employ a model of laser radiation in which  $I_{ph} = 0$ , and  $Q(I_{QP})$  is determined by the laser intensity.

#### 1. Steady-state response to spatially uniform cw radiation

When the applied fields are constant for all of space and time, i.e.,  $I_{QP}$  and  $I_{ph}$  are constants, the left-hand side of Eq. (2.17a) is equal to zero, and the resulting solution for  $n_{QP}$ , now labeled  $n_{QP}^{SS}$ , is

$$n_{QP}^{SS} = n_{QP}^0 (1 + Q)^{1/2}, \quad (2.19)$$

which when used in Eq. (2.17b) gives

$$n_{ph} = \frac{n_{ph}^0}{(1 + \tau_{es}/\tau_B)} + I_{ph} \tau_{ph} + n_{QP}^0 \frac{\tau_{ph}}{2\tau_R} (1 + Q). \quad (2.20)$$

Here  $Q$  is a dimensionless drive current given by

$$Q = (1 + \tau_{es}/\tau_B) \tau_R^0 \left[ \frac{I_{QP}}{n_{QP}^0} + \frac{2\tau_{es}}{\tau_{es} + \tau_B} \frac{I_{ph}}{n_{QP}^0} \right]. \quad (2.21)$$

In the limit of weak external fields, i.e.,  $Q \ll 1$ , we see that to first order

$$\delta n_{QP} \equiv n_{QP}^{SS} - n_{QP}^0 = n_{QP}^0 \frac{Q}{2}. \quad (2.22)$$

In the opposite limit,  $Q \gg 1$ , for strong external fields driving the superconductor far from equilibrium, we see that

$$\delta n_{QP} = n_{QP}^0 Q^{1/2}. \quad (2.23)$$

#### 2. Transient response to spatially uniform pulsed radiation

We shall consider two cases for which the amplitudes of the external driving fields (and, hence, the driving currents  $I$ ) vary with time, but are still homogeneous in space.

*a. Sudden onset and the approach to steady state.* In this case the laser is turned on to a constant value at  $t=0$ :  $I_{QP(ph)} = I_{QP(ph)}^0 \Theta(t)$ . Initially the system is in equilibrium and must approach the steady-state value at  $t = \infty$ . It is convenient to introduce the dimensionless quantities:  $u = n_{QP}/n_{QP}^{SS}$ , from Eq. (2.3a), and

$$x = t/\bar{\tau}_R, \quad (2.24)$$

where

$$\bar{\tau}_R = \tau_R (1 + \tau_{es}/\tau_B). \quad (2.25)$$

This  $\bar{\tau}_R$ , the time scale over which the QP density will

vary, is determined by the recombination time (steady state), increased by the well-known factor  $(1 + \tau_{es}/\tau_B)$ , which arises from phonon trapping. With these definitions, the QP equation (2.17a) becomes

$$\frac{du}{dx} = \Lambda(x) - \left[ u^2 - \left[ \frac{n_{QP}^0}{n_{QP}^{SS}} \right]^2 \right], \quad (2.26)$$

where

$$\Lambda(x) = \bar{\tau}_R \left[ I_{QP}(x) + \frac{2\tau_{es}}{\tau_{es} + \tau_B} I_{ph}(x) \right] \frac{1}{n_{QP}^{SS}}, \quad (2.27)$$

which for the step-function drive, now under consideration becomes

$$\Lambda(x) = \Lambda^0 \Theta(x) = \bar{\tau}_R \left[ I_{QP}^0 + \frac{2\tau_{es}}{\tau_{es} + \tau_B} I_{ph}^0 \right] \frac{1}{n_{QP}^{SS}} \Theta(x). \quad (2.28)$$

Now, as  $t \rightarrow \infty$ , meaning  $x \rightarrow \infty$ , we have  $u = 1$  and  $du/dx = 0$ . Putting these limiting values into Eq. (2.26), with  $\Lambda(x) = \Lambda^0$  we find that

$$\Lambda^0 = 1 - \left[ \frac{n_{QP}^0}{n_{QP}^{SS}} \right]^2, \quad (2.29)$$

which put back into Eq. (2.26) yields the equation, for  $x > 0$ ,

$$\frac{du}{dx} = 1 - u^2, \quad (2.30)$$

with the initial condition  $u(0) = n_{QP}^0/n_{QP}^{SS}$ .

The solution of this first-order nonlinear Riccati equation is found in standard tables.<sup>9</sup> We apply the boundary condition; multiply by  $n_{QP}^{SS}/n_{QP}^0$ , in order to normalize with respect to the equilibrium values; and, finally, arrive at the solution

$$\frac{n_{QP}(t)}{n_{QP}^0} = \sqrt{1+Q} \left[ \frac{\cosh(t/\bar{\tau}_R) + \sqrt{1+Q} \sinh(t/\bar{\tau}_R)}{\sinh(t/\bar{\tau}_R) + \sqrt{1+Q} \cosh(t/\bar{\tau}_R)} \right], \quad (2.31)$$

where the  $I_{QP(ph)}$  in the definition of  $Q$  are understood to be  $I_{QP(ph)}^0$ .

*b. Pulsed laser.* If the laser is turned on at  $t=0$  and off at  $t=t_0$ :

$$I_{QP(ph)}(t) = I_{QP(ph)}^0 \Theta(t) \Theta(t_0 - t),$$

then the system is initially in equilibrium and as  $t \rightarrow \infty$  it returns to equilibrium. We, therefore, define the dimensionless variables with respect to their equilibrium values:  $u = n_{QP}/n_{QP}^0$ , from Eq. (2.3b), and

$$x = t/\bar{\tau}_R^0, \quad (2.32)$$

where

$$\bar{\tau}_R^0 = \tau_R^0 (1 + \tau_{es}/\tau_B) \quad (2.33)$$

is the enhanced recombination time (for equilibrium). Furthermore, we take the definition of  $\Lambda^0$  from Eq. (2.28),

but replace  $n_{QP}^{SS}$  by  $n_{QP}^0$  and  $\tau_R$  by  $\tau_R^0$ , thus making  $\Lambda^0 = Q$  for  $Q$  as defined by Eq. (2.21) but with  $I_{QP(ph)}$  understood to be  $I_{QP(ph)}^0$ . Using these definitions, we find that Eq. (2.17a) for the QP becomes

$$\frac{du}{dx} = \Lambda^0 \Theta(x) \Theta(x_0 - x) - (u^2 - 1). \quad (2.34)$$

We solve this equation for  $0 \leq x \leq x_0$  ( $0 \leq t \leq t_0$ ) subject to the boundary condition  $u(0) = 1$ , and we then solve for  $x_0 \leq x$  subject to the condition that the solution  $u$  be continuous at  $x = x_0$ .

The solution, for  $0 < t < t_0$ , is

$$\frac{n_{QP}(t)}{n_{QP}^0} = a \left[ \frac{\cosh(at/\tau_R^0) + a \sinh(at/\tau_R^0)}{\sinh(at/\tau_R^0) + a \cosh(at/\tau_R^0)} \right], \quad (2.35)$$

where

$$a = (1 + \Lambda^0)^{1/2} = (1 + Q)^{1/2} = \frac{n_{QP}^{SS}}{n_{QP}^0}. \quad (2.36)$$

With Eqs. (2.5a), (2.5b), (2.25), and (2.33), we see that

$$\frac{\tau_R^0}{a} = \tilde{\tau}_R. \quad (2.37)$$

Thus, Eq. (2.35), describing  $n_{QP}(t)$  after the external fields are turned on, but before they are turned off, is identical to Eq. (2.31) which describes the sudden switching on.

The solution for  $t > t_0$  is

$$\frac{n_{QP}(t)}{n_{QP}^0} = \frac{n_0 \cosh[(t-t_0)/\tau_R^0] + \sinh[(t-t_0)/\tau_R^0]}{n_0 \sinh[(t-t_0)/\tau_R^0] + \cosh[(t-t_0)/\tau_R^0]}, \quad (2.38)$$

where  $n_0$  is the value of  $n_{QP}(t_0)$  evaluated from Eq. (2.35).

These results show that when a field consisting of a single pulse is turned on, the QP density starts to rise (heading toward the steady-state value), with a rise-time constant of  $\tilde{\tau}_R$ ; and when the field is shut off, the density decays back toward its equilibrium value with a decay-time constant of  $\tau_R^0$ . These results are demonstrated graphically in Fig. 1. This figure shows the results of Eq. (2.31) for the sudden switching on of the fields, to a constant value, and of Eqs. (2.35)–(2.38) for a pulsed field with two different pulse lengths  $t_0$ . All times have been normalized with respect to the same rise time  $\tilde{\tau}_R$ . The strength of the fields, when switched on, is the same in all cases, as determined by the value of the dimensionless parameter  $Q$ , here taken to be  $Q = 73$ . The significance of this value of  $Q$  and the range of possible values of  $\tilde{\tau}_R$  are discussed in Sec. II C.

### 3. Response to a weak oscillating electromagnetic grating

We now let the external driving currents vary in both time and space to describe, for example, the response to two laser sources that create an electromagnetic grating. For two laser beams described by wave vectors, frequen-

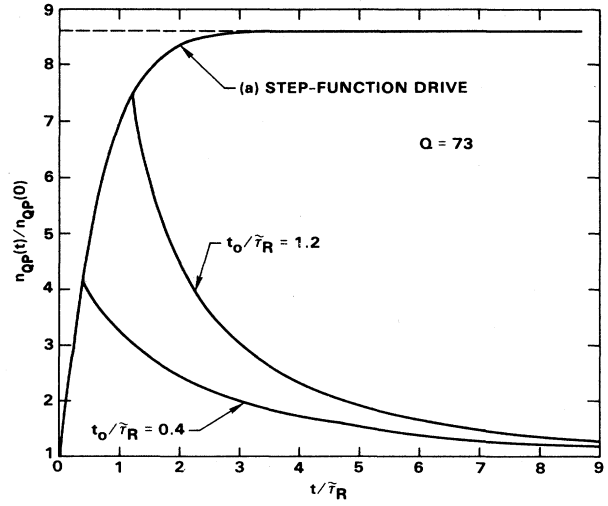


FIG. 1. Evolution of the nonequilibrium quasiparticle density (with respect to its equilibrium value) as a function of time [normalized to the rise-time constant (to steady state),  $\tilde{\tau}_R$ ]. Curve (a): Approach to steady state, for sudden switching on at  $t = 0$  of drive current to a constant value ( $Q$ ). Lower two curves: Response to a time-pulse—on at  $t = 0$ , off at  $t = t_0$ , and constant  $Q$  at  $0 < t < t_0$ .

cies, and polarizations ( $\mathbf{K}_1, \omega_1, \hat{\mathbf{e}}_1$ ) and ( $\mathbf{K}_2, \omega_2, \hat{\mathbf{e}}_2$ ), the total light intensity at the superconductor's surface is

$$S(\mathbf{r}, t) = S_1 + S_2 + 2(\zeta S_1 S_2)^{1/2} \cos(\mathbf{K} \cdot \mathbf{r} - \Omega t), \quad (2.39)$$

where  $S_i$  is the intensity of laser  $i$ ;  $\mathbf{K} = \mathbf{K}_1 - \mathbf{K}_2$ ;  $\Omega = \omega_1 - \omega_2$ ; and  $\zeta = (\hat{\mathbf{e}}_1 \cdot \hat{\mathbf{e}}_2)^2$ . The last term represents the grating, which creates a quasiparticle current that oscillates with the same  $\mathbf{K}$  and  $\Omega$ . An electromagnetic grating of the form

$$(\mathbf{K}_1 + \mathbf{K}_2) \cdot \mathbf{r} - (\omega_1 + \omega_2)t$$

is also created, but for visible or near-infrared lasers its time scale is too fast for either the QP or phonons to respond. These considerations motivate us to study the QP response to the laser induced current

$$I_{QP}(\mathbf{r}, t) = [I_0 + \Delta I \cos(\mathbf{K} \cdot \mathbf{r} - \Omega t)] \Theta(t), \quad (2.40)$$

where  $I_0 = I_1 + I_2$ ,  $\Delta I = (\zeta I_1 I_2)^{1/2}$ , and where again through the  $\Theta$  function we shall include the transient effects associated with turning on the laser at  $t = 0$ . We shall consider two special cases: (1) both  $I_1$  and  $I_2$  are weak, so that  $I_0$  and  $\Delta I$  are weak, and the departure from equilibrium is small, and (2)  $I_1$  and  $I_2$  may be strong but  $\Delta I$  is weak (experimentally realizable by using nearly orthogonal beams).

In the first case, when both beams are weak, we keep terms only to first order in the departure from equilibrium:

$$\delta n_{QP}(\mathbf{r}, t) = n_{QP}(\mathbf{r}, t) - n_{QP}^0,$$

which yields a linearized version of Eq. (2.17a),

$$\left[ \frac{\partial}{\partial t} - D_{\text{QP}} \nabla^2 \right] \delta n_{\text{QP}} = \left[ I_{\text{QP}} + \frac{2\tau_{\text{es}}}{\tau_B + \tau_{\text{es}}} I_{\text{ph}} \right] - \frac{2}{\tau_R^0 (1 + \tau_{\text{es}}/\tau_B)} \delta n_{\text{QP}}, \quad (2.41)$$

where now  $I_{\text{ph}}=0$  and  $I_{\text{QP}}$  is given by Eq. (2.40). Since this equation is linear in  $\delta n_{\text{QP}}$ , the solution is a linear combination,  $\delta n_{\text{QP}}^0 + \delta n_{\text{QP}}^1$ , of the responses to the  $I_0$  term and to the  $\Delta I$  term in Eq. (2.40). The former is given already by Eq. (2.31) and the latter is given by

$$\delta n_{\text{QP}}^1 = \frac{\tau \Delta I e^{i(\mathbf{K} \cdot \mathbf{r} + \phi)}}{2[1 + (\Omega\tau)^2]^{1/2}} [e^{-i\Omega t} - e^{-t/\tau}] + \text{c.c.}, \quad (2.42)$$

where

$$\tau^{-1} = D_{\text{QP}} K^2 + \frac{2}{\tau_R^0 (1 + \tau_{\text{es}}/\tau_B)} \quad (2.43)$$

and

$$\phi = \tan^{-1} \Omega \tau. \quad (2.44)$$

The transient response, given by the second exponential in the brackets in Eq. (2.42), is characterized by the new response time  $\tau$ , which is now set by the effective grating-diffusion time,  $(D_{\text{QP}} K^2)^{-1}$ , as well as by the enhanced recombination time,  $\tau_R^0$ . We can estimate which of the two terms dominates by using the classical approximation  $D_{\text{QP}} = \frac{1}{3} v_F^2 \tau_R^0$ . Thus, the ratio of the two contributions (diffusion:recombination) becomes

$\frac{2}{3}(v_F K \tau_R^0)^2$ : the diffusion term dominates if  $v_F K \tau_R^0 \gg 1$  (i.e., if a QP diffuses a distance greater than a wavelength of the driving field before recombining), and the QP recombination dominates if  $v_F K \tau_R^0 \ll 1$ . If we estimate  $v_F$  as  $\cong 10^8$  cm/s and  $\tau_R^0$  anywhere from  $10^{-10}$  to  $10^{-7}$  s, then this ratio has an order-of-magnitude value from  $10^{-4} K^2$  to  $10^2 K^2$ . The wave vector difference,  $K$ , from interfering laser beams can be varied continuously. If  $K \gg 1 \times 10^2$  cm $^{-1}$ , the diffusion term dominates the transient response.

We note that for weak drive currents of *arbitrary* spatial dependence, the solution of the linearized equation for  $\delta n_{\text{QP}}$  is found by summing the result in Eq. (2.42), for all Fourier components  $\Delta I(\mathbf{K}, \Omega)$  of the current, over  $\mathbf{K}$ .

For the case where the two laser intensities are strong but  $\Delta I$  is weak, we return to the nonlinear differential equation, Eq. (2.17a) [with  $I_{\text{ph}}=0$  and  $I_{\text{QP}}$  given by Eq. (2.40)], and we say that  $n_{\text{QP}} = n_1(t) + \delta n(\mathbf{r}, t)$ . Here,  $n_1$  is the response to  $I_0$ , which again is given by Eq. (2.31); and  $\delta n(\mathbf{r}, t)$  ( $\ll n_1$ ) is given by the equation

$$\left[ \frac{\partial}{\partial t} - D_{\text{QP}} \nabla^2 \right] \delta n(\mathbf{r}, t) = \Delta I \cos(\mathbf{K} \cdot \mathbf{r} - \Omega t) - \frac{2}{\tau_R^1(t)} \delta n(\mathbf{r}, t), \quad (2.45)$$

where

$$\tau_R^1(t) = [2Rn_1(t)]^{-1} (1 + \tau_{\text{es}}/\tau_B)$$

is the instantaneous recombination time in the presence of the QP drive current  $I_0$ . The solution to Eq. (45) is

$$\delta n(\mathbf{r}, t) = \frac{1}{2} \Delta I e^{i\mathbf{K} \cdot \mathbf{r}} \exp \left[ - \int_0^t dt' \frac{1}{\tau(t')} \right] \int_0^t dt' e^{-i\Omega t'} \exp \left[ + \int_0^{t'} dt'' \frac{1}{\tau(t'')} \right] + \text{c.c.}, \quad (2.46)$$

where the QP response time  $\tau(t)$  is given by

$$\tau(t)^{-1} = D_{\text{QP}} K^2 + \frac{2}{\tau_R^1(t)}. \quad (2.47)$$

### C. Magnitude of laser drive current $Q$ and response time $\tilde{\tau}_R$

The preceding results show that the departure from equilibrium, both steady-state and time-varying, is governed by the value of  $Q$ , on a time scale set by  $\tilde{\tau}_R$ . We now estimate the values of these constants. The numerical results that will be presented in the following sections will all be given in terms of  $Q$  (or  $Q'$  defined below) and  $\tilde{\tau}_R$ . For real practical applications, we wish to know the numerical values of these parameters in terms of real laser powers and real time scales.

In estimating the magnitude of the dimensionless drive current  $Q$ , we first notice that this quantity, as defined through Eq. (2.21), is highly temperature dependent. Systems at different temperatures but characterized by the same value of  $Q$  correspond to different driving powers,

e.g., different laser intensities. To compare systems at different temperatures but with the same external drive, we would like to have a parameter that characterizes the external drive strength independent of temperature. Now,  $Q$  depends on  $T$  through the equilibrium QP density  $n_{\text{QP}}^0$  and recombination time  $\tau_R^0$ . The pair-breaking time  $\tau_B$  also depends on the temperature, but only weakly for  $T < 0.5T_c$ . In any case,  $\tau_B$  appears in  $Q$  only through the ratio  $\tau_B/\tau_{\text{es}}$ ; and since  $\tau_{\text{es}}$  [being dependent on the film's thickness and coupling to the reservoir (substrate)] is an adjustable macroscopic parameter, it is usual in the present sort of phenomenology to take the ratio  $\tau_B/\tau_{\text{es}}$  itself as the adjustable parameter.<sup>5,6</sup> Taking  $\tau_B/\tau_{\text{es}}$  as a given fixed quantity, we have the temperature dependence of  $Q$  only through  $n_{\text{QP}}^0$  and  $\tau_R^0$ , and thus we can define a new dimensionless drive current  $Q'$  which is temperature independent:

$$Q' = \gamma Q, \quad (2.48)$$

where

$$\gamma = 2n^0 / (\tau_R^0 / \tau^0), \quad (2.49)$$

with  $n^0$  defined as the usual normalized QP density,

$$n^0 = n_{\text{QP}}^0 / 4N(0)\Delta(0). \quad (2.50)$$

Here,  $N(0)$  is the electron density of states at the Fermi energy in the normal state,  $\Delta(0)$  is the zero-temperature gap energy, and  $\tau^0$  is a temperature-independent scale factor (see Ref. 6). If we employ the definition of  $\tau_R^0$ , Eq. (2.5b), we find that

$$\gamma = \left[ \frac{R\tau^0}{N(0)\Delta(0)} \right] (n_{\text{QP}}^0)^2. \quad (2.51)$$

The temperature dependence of  $\gamma$ , and hence of  $Q$ , arises from the temperature dependence of  $(n_{\text{QP}}^0)^2$ .

To calculate  $\gamma(T)$ , we use Eq. (2.49), and for  $\tau_R^0 / \tau^0$  we employ the expression found by Kaplan *et al.* [Ref. 10, Eq. (11)] for a QP at the gap edge (for  $T \ll T_c$ , the leading term in this expression for  $\tau_R / \tau^0$  is proportional to  $\exp[\Delta(0)/kT]$ ). For  $n_{\text{QP}}^0$  we use the BCS expression [see Eq. (3.2)]. The resulting temperature dependence of the factor  $\gamma$  is plotted in Fig. 2 for Al. For a given set of drive currents,  $I_{\text{QP}}$  and  $I_{\text{ph}}$ , we can calculate  $Q'$ , and then from  $\gamma(T)$  we can determine  $Q$  at any temperature  $T$ :

$$S = (45 \text{ mW/cm}^2)(1 + \tau_{\text{es}}/\tau_B)^{-1} Q' \approx (0.45 - 23 \text{ mW/cm}^2) Q', \quad \text{for Al}, \quad (2.53a)$$

$$S = (2 \text{ kW/cm}^2)(1 + \tau_{\text{es}}/\tau_B)^{-1} Q' \approx (1 - 100 \text{ W/cm}^2) Q', \quad \text{for Nb}, \quad (2.53b)$$

where we have taken

$$\tau_{\text{es}}/\tau_B = (4d/\eta v_{\text{ph}})/\tau_B$$

as  $\approx 1/\eta$  for Al and  $\approx 20/\eta$  for Nb, with  $1/\eta = 1-100$ . The values of  $Q'$  on the order of unity, which are used for the numerical results presented here, thus correspond to a few mW/cm<sup>2</sup> on Al or a few W/cm<sup>2</sup> on Nb. If  $Q' = 1$ , then  $Q = \gamma^{-1}$ , which for Al equals 19 at  $0.5T_c$  and equals

$$\bar{\tau}_R \equiv \tau_R^0 (1 + \tau_{\text{es}}/\tau_B) (1 + \gamma^{-1} Q')^{-1/2} \approx \tau_R^0 (1 + \tau_{\text{es}}/\tau_B) (\gamma^{-1} Q')^{-1/2}, \quad \text{for } Q \gg 1,$$

is independent of temperature, since  $\tau_R^0 \gamma^{1/2}$  is  $T$  independent; inversely proportional to the square root of the incident power; and highly material dependent, through  $\tau_R^0$ . For the case of  $Q = 1.6 \times 10^5$  and  $T = 0.25T_c$  considered later for Nb ( $Q' = 3$ )

$$\bar{\tau}_R = (18 \text{ ps})(1 + \tau_{\text{es}}/\tau_B),$$

which for a 1- $\mu\text{m}$ -thick film becomes  $\bar{\tau}_R = 0.38-36 \text{ ns}$  (as  $\eta^{-1} = 1-100$ ). Thus, rise times as fast as nanoseconds may be possible in this range of parameters.

Whereas the rise time is temperature independent, the recovery time to equilibrium after a light pulse has

$Q = \gamma^{-1}(T)Q'$ . The increase by orders of magnitude in  $\gamma^{-1}$  as the temperature is lowered indicates that a nonlinear response ( $Q \gg 1$ ) may be hard to avoid.

For an optically driven superconductor we express  $Q'$  as a function of the light intensity following Parker's analysis.<sup>7</sup> The fraction of the absorbed energy resulting in QP creation is  $F \approx 1$  and the average energy per QP is  $\approx \Delta(T)$ . Thus,  $I_{\text{QP}} \approx PF/\Delta(T)V$ , where  $P$  is the power absorbed in volume  $V$ ; and we take  $I_{\text{ph}} = 0$ . The  $\Delta(T)$  puts some  $T$  dependence into  $Q'$ , which is avoided by redefining  $\gamma$  by multiplying the right-hand side of Eq. (2.49) by  $\Delta(T)/\Delta(0)$ . Since in the temperature ranges that concern us, we have  $\Delta(T)/\Delta(0) \approx 1$ , this modifying factor has no real effect on  $\gamma(T)$  for the present estimates. If we say that QP are created uniformly down to a distance  $\bar{d}$ , which is the film's thickness  $d$  or the penetration depth, whichever is smaller, then we have

$$Q'(S) = \left[ \frac{F\tau^0(1 + \tau_{\text{es}}/\tau_B)}{2N(0)\Delta^2(0)} \right] \frac{\mathcal{A}}{\bar{d}} S, \quad (2.52)$$

where  $S$  is the power flux per unit area in the incident laser beam, and  $\mathcal{A}$  is the film's absorptance. If we take  $F \approx 1$  and use low-temperature optical data for  $\mathcal{A}$  and  $\bar{d}$  for 1- $\mu\text{m}$  incident radiation for a 1- $\mu\text{m}$ -thick film, we arrive at the following estimates:

$5.32 \times 10^4$  at  $0.25T_c$ —well into the nonlinear regime. To maintain a linear response ( $Q < 1$ ) for Al requires driving powers of less than 0.1 mW/cm<sup>2</sup> at  $0.5T_c$  and less than  $10^{-4}$  mW/cm<sup>2</sup> at  $0.25T_c$ . Our results to be presented for Nb, at  $Q' = 3$  and  $T = 0.25T_c$ , correspond to  $Q = 1.6 \times 10^5$ , again in the nonlinear regime. At  $0.25T_c$ , only laser powers of  $\mu\text{W/cm}^2$  on Nb will give a linear response.

The rise time to steady state

passed,

$$\bar{\tau}_R^0 = \tau_R^0 (1 + \tau_{\text{es}}/\tau_B)$$

depends on temperature, as the QP recombination depends on  $n_{\text{QP}}^2$ . In the example where  $Q = 1.6 \times 10^5$  at  $T = 0.25T_c$ ,

$$\bar{\tau}_R^0 = 400\bar{\tau}_R = 0.15-14 \mu\text{s}.$$

If speed is a requirement, for example, nanosecond response, we must go to higher temperatures; at  $T = 0.5T_c$  the recovery is faster by a factor of 40.

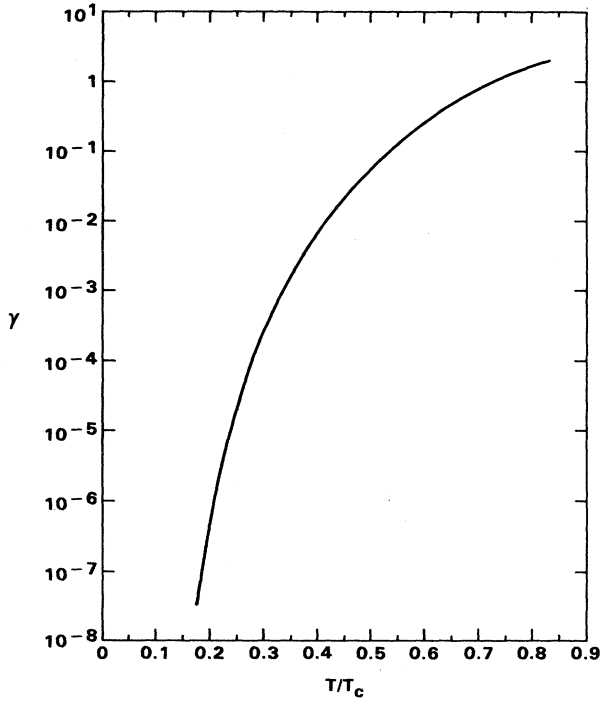


FIG. 2. The dimensionless scale factor  $\gamma$ , which contains the temperature dependence of the effective drive current  $Q(T)$ , vs the reduced temperature  $T/T_c$ .

### III. STATISTICAL MECHANICS IN THE $T^*$ APPROACH

In Sec. II we have calculated the total QP density  $n_{QP}(\mathbf{r}, t)$  for various external driving fields. What is required, however, in order to determine the electrodynamic response of the system, is knowledge of the energy distribution of the QP density. For the case in which a superconductor is driven from equilibrium by a strong electromagnetic field at optical frequencies, and then is probed by a weak microwave field, the well-known  $T^*$  model of Parker has been shown to be effective.<sup>7,11</sup>

It is assumed in this model that the optically excited QP come into equilibrium with the phonons of energy greater than  $2\Delta$ , at an effective temperature  $T^*$ . The value of  $T^*$  is determined by the BCS theory to correspond to the value of  $n_{QP}$ . We briefly outline the equations that we employ in implementing the Parker model, since these equations will serve in the later discussion.

We wish to look at systems driven far from equilibrium, when many QP are created, for the purpose of exploring departures from the linear response to the driving fields. Thus, at this point, we avoid using any linearized equations, and we employ the exact relation between  $n_{QP}$  and  $\Delta$ ,

$$n_{QP} = 4N(0) \int_{\Delta}^{\infty} \frac{EdE}{(E^2 - \Delta^2)^{1/2} (e^{E/kT} + 1)}. \quad (3.1)$$

In Eq. (3.1), if  $n_{QP}$  is the equilibrium density,  $n_{QP}^0$ , then  $T$  is the ambient temperature and  $\Delta$  is the equilibrium gap

$\Delta(T)$ ; but if  $n_{QP}$  is the nonequilibrium density, then  $T$  is replaced by the effective temperature  $T^*$  and  $\Delta$  is the nonequilibrium gap  $\Delta(T^*)$ . With the weak-coupling result that  $\Delta(0) = 1.764kT_c$ , the integral in Eq. (3.1) can be transformed to give the results

$$n_{QP}^0 = 4N(0)\Delta(0)I(\beta, T), \quad (3.2a)$$

$$n_{QP} = 4N(0)\Delta(0)I(\beta, T^*), \quad (3.2b)$$

where

$$\beta = \frac{\Delta(T)}{kT} \quad \text{and} \quad \beta^* = \frac{\Delta(T^*)}{kT^*}, \quad (3.3)$$

$$T = \frac{T}{T_c} \quad \text{and} \quad T^* = \frac{T^*}{T_c}, \quad (3.4)$$

and where

$$I(\beta, T) = \int_0^{\infty} dy \{1 + \exp[\beta^2 + (1.764/T)y^2]^{1/2}\}^{-1}. \quad (3.5)$$

One of the two variables  $\beta$  and  $T$ , which are arguments on  $I$ , can be eliminated by use of the BCS gap equation, from which we find, in the case of weak coupling,

$$T = T(\beta) = \frac{1.764}{\beta} e^{F(\beta)}, \quad (3.6)$$

or, for  $T^*$  and  $\Delta(T^*)$ ,  $T^* = T(\beta^*)$ , where

$$F(\beta) = \frac{-\beta}{2} \int_0^{\infty} \frac{x \sinh^{-1} x \operatorname{sech}^2([1+x^2]^{1/2}\beta/2)}{(1+x^2)^{1/2}} dx. \quad (3.7)$$

The RT equations give us a normalized QP density  $n_{QP}/n_{QP}^0$ , which the two equations (3.2a) and (3.2b) allow us to express as

$$\frac{n_{QP}}{n_{QP}^0} = \frac{I(\beta^*, T(\beta^*))}{I(\beta, T)}. \quad (3.8)$$

We know the left-hand side of Eq. (3.8) and also the denominator on the right-hand side. Therefore,  $\beta^*$  is the only unknown, and is found numerically. After we have found  $\beta^*$ , we substitute it back into Eq. (3.6) to find  $T^*/T_c$ , and substitute it into the gap equation to find the nonequilibrium gap  $\Delta$ :

$$\frac{\Delta}{\Delta(0)} = \frac{\Delta(T^*)}{\Delta(0)} = e^{F(\beta^*)}. \quad (3.9)$$

Finally, the distribution function for the nonequilibrium case  $f^*(E)$ , is found by evaluating the Fermi function  $f_0(E, T)$  at  $T^*$ :  $f^*(E) = f_0(E, T^*)$ .

When the physical quantities pertaining to the nonequilibrium superconducting state are properly normalized [e.g.,  $\delta\Delta/\Delta(T)$  and  $f^*(\bar{E})$ , where  $\bar{E} = E/\Delta(T)$ ], they are all material independent.

When  $n_{QP}$  on the left-hand side of Eq. (3.1) or (3.8) is a function of " $t$ ," we can solve for  $\beta^*$  (as well as  $\Delta$  and  $f$ ) as a function " $t$ ." This time-dependent  $T^*$  approach is legitimate when the time-scale for the changing drive current (for example, the pulse width of the square-wave drive



due to a pulsed laser) is long compared to the time for the QP to relax to the bottom of the conduction band and Eq. (2.18) is satisfied. This statement follows from the fact that detailed balance implies that the QP and the phonons are in equilibrium with one another. We shall use the time-dependent kinetics in Sec. IV to obtain the time-dependent conductance and thin-film transmittance.

#### IV. ELECTRODYNAMICS

We treat the electrodynamic response of a superconducting thin film by use of the Mattis-Bardeen (MB) equations.<sup>8</sup> The MB equations give the ac conductivity of the superconductor, with respect to its normal-state conductivity:

$$\sigma/\sigma_n = (\sigma_1 - i\sigma_2)/\sigma_n.$$

These equations, valid in the extreme local (dirty) limit and in the extreme anomalous limit, have long been applied successfully to treat thin films.<sup>12,13</sup>

In Sec. IV A, we shall use the  $T^*$  model from Sec. III together with the MB equations in an exact formulation. In Sec. IV B we shall develop a perturbative approach which, using the MB equations, expresses the electrodynamic response functions to first order in  $\delta\Delta$ .

##### A. Exact formulation

The MB expression for the conductivity employs the QP distribution function  $f$  and the energy gap  $\Delta$ . Again, following Parker's model, we can obtain the nonequilibrium value for  $\sigma/\sigma_n$  by using the  $T^*$  values:  $f_0(E, T^*)$  and  $\Delta(T^*)$ . The MB equations for  $\sigma_1/\sigma_n$  and  $\sigma_2/\sigma_n$  can be cast as explicit functions of  $\beta$ , as defined in Eqs. (3.3), and of  $\mathcal{E} = \hbar\omega/\Delta(T)$ . With the definition that  $\mathcal{E}^* = \hbar\omega/\Delta(T^*)$ , the change in conductivity, i.e., the difference between the nonequilibrium and equilibrium values, is expressed as

$$\delta\sigma_i/\sigma_n = \sigma_i(\mathcal{E}^*, \beta^*)/\sigma_n - \sigma_i(\mathcal{E}, \beta)/\sigma_n, \quad \text{for } i=1 \text{ or } 2. \quad (4.1)$$

When we plot  $\delta\sigma/\sigma_n$  (or an electrodynamic function thereof) versus  $Q$ , and repeat this for different temperatures, the results are material independent, but a given value of  $Q$  on the different  $T$  curves does not correspond to a given driving power. When we plot the electrodynamic versus  $Q'$ , however, the results are material dependent (via  $\gamma$ ), but a given value of  $Q'$  on different  $T$  curves corresponds to a given power. The same is true for the superconducting parameters [ $\delta\Delta/\Delta(T)$ ,  $\delta f$ , and  $\delta T/T$ ] themselves, from which the electrodynamic results follow.

Our steady-state results for  $\delta\sigma_1/\sigma_1$  and  $\delta\sigma_2/\sigma_2$  are shown in Fig. 3. The fractional change in the real part of  $\sigma$  is seen to be much greater than in the imaginary part. The real part is determined by  $n_{QP}$ , and there is a large fractional change in this quantity due to pair breaking by the optical drive. The imaginary part, on the other hand, is mostly determined by the pair density, which under-

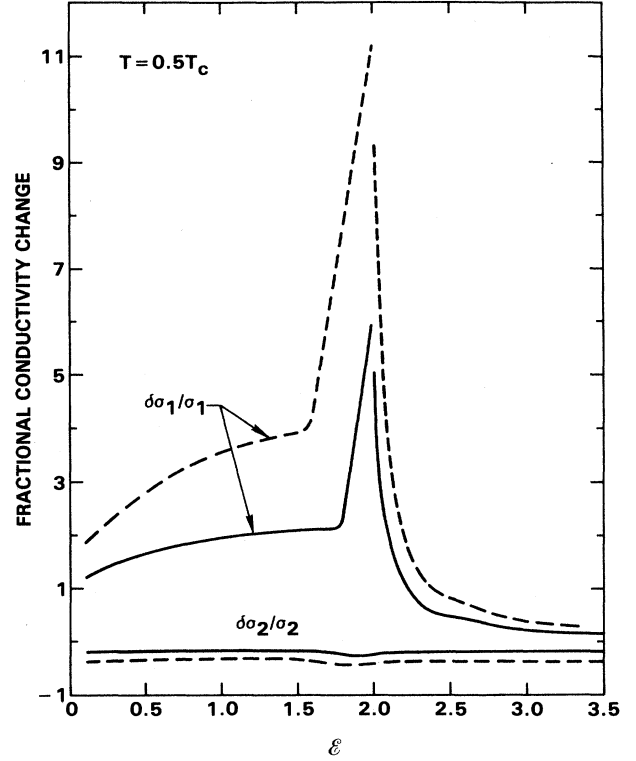


FIG. 3. The fractional change in the (complex) conductivity, between equilibrium and steady state, for a superconducting thin film vs the probe frequency, expressed as the photon energy divided by the equilibrium gap energy  $\mathcal{E} = \hbar\omega/\Delta(T)$ . The upper two curves show the real part  $\delta\sigma_1/\sigma_1$  and the lower two curves the imaginary part  $\delta\sigma_2/\sigma_2$ . At ambient temperature  $T = 0.5T_c$ . Dashed curves: for an external effective drive current of  $Q = 30$  ( $Q' = 1.597$  for Al). Solid curves: for  $Q = 10$  ( $Q' = 0.532$ ).

goes only small fractional changes. The largest changes are seen for frequencies just below the gap, reflecting the reduction in the gap,  $\delta\Delta$ .

The transient response to pulsed-laser radiation can be obtained from the solutions of the RT equations, given by Eqs. (2.35) and (2.38). Figure 4 depicts the time evolution of the fractional change in the real part of the conductivity for a pulse of duration  $t_0 = 1.2\bar{\tau}_R$  for  $Q = 1.6 \times 10^5$  at  $T = 0.25T_c$  (for Nb this means  $Q' = 3$ ). The three curves correspond, in the case of Nb, to the three frequencies 17, 60, and 240 GHz. The fractional change in  $\sigma_1$  is greater at the shortest wavelengths, reflecting the role of the QP density in  $\sigma_1$ . A comparison of  $\delta\sigma_1/\sigma_1$  to  $\delta n_{QP}/n_{QP}^0$  shows that the transient behavior of these quantities is quite similar.

To describe the transmittance  $t_s$  of a superconducting thin film on a dielectric substrate, we use the simplified analysis of Glover and Tinkham,<sup>14</sup> in which we neglect edge effects and multiple reflections in the substrate. In the normal state the transmittance is

$$t_N = [1 + Z_0/(\epsilon + 1)R_N]^{-2},$$

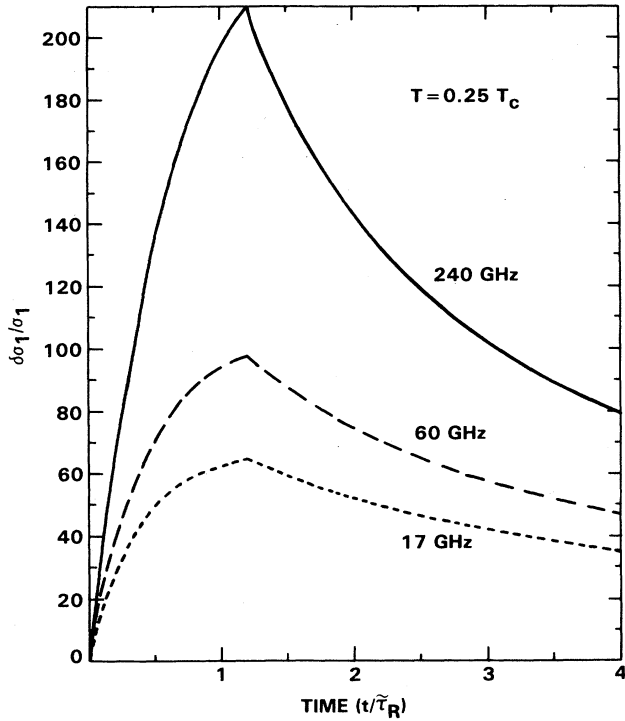


FIG. 4. Fractional change in the real part of the conductivity vs time, for a light pulse of duration  $t_0 = 1.2\tilde{\tau}_R$  and strength  $Q = 1.6 \times 10^5$  at  $T = 0.25T_c$  ( $Q' = 3$  for Nb). For frequencies which in Nb correspond to 240 GHz (solid), 60 GHz (long-dashed line), and 17 GHz (short-dashed line).

where  $R_N (= 1/\sigma_n d)$  is the dc resistance per square of the film,  $Z_0$  is the impedance of free space,  $d$  the film thickness, and  $n$  the index of refraction for the substrate. Evaluating the  $\sigma_1$  in the expression for  $t_s$  at  $\mathcal{E}^*, T^*$  and then again at  $\mathcal{E}, T$  and subtracting the results, we find

$$\frac{\delta t_s}{t_s} = \frac{[(t_N^{-1/2} - 1)^{-1} + \sigma_1/\sigma_n]^2 + (\sigma_2/\sigma_n)^2}{[(t_N^{-1/2} - 1)^{-1} + \sigma_1^*/\sigma_n]^2 + (\sigma_2^*/\sigma_n)^2} - 1, \quad (4.2)$$

where  $\sigma_i = \sigma_i(\mathcal{E}, \beta)$  and  $\sigma_i^* = \sigma_i(\mathcal{E}^*, \beta^*)$ . Similarly,  $\delta t_s/t_N$  is found by evaluating the  $\sigma_i$  in the following equation

$$\frac{t_s}{t_N} = \{ [t_N^{1/2} + (1 - t_N^{1/2})(\sigma_1/\sigma_n)]^2 + [(1 - t_N^{1/2})(\sigma_2/\sigma_n)]^2 \}^{-1}, \quad (4.3)$$

first at  $\mathcal{E}^*, T^*$  and then at  $\mathcal{E}, T$  and subtracting the two. These results demonstrate that the quantities  $\delta t_s/t_N$  and  $\delta t_s/t_s$  both depend on the film thickness and material properties only through the  $t_N$ , and this quantity is a function of only the resistance  $R_N$  and substrate index of refraction  $n$ .

Figure 5 demonstrates the results for  $\delta t_s/t_N$  versus the

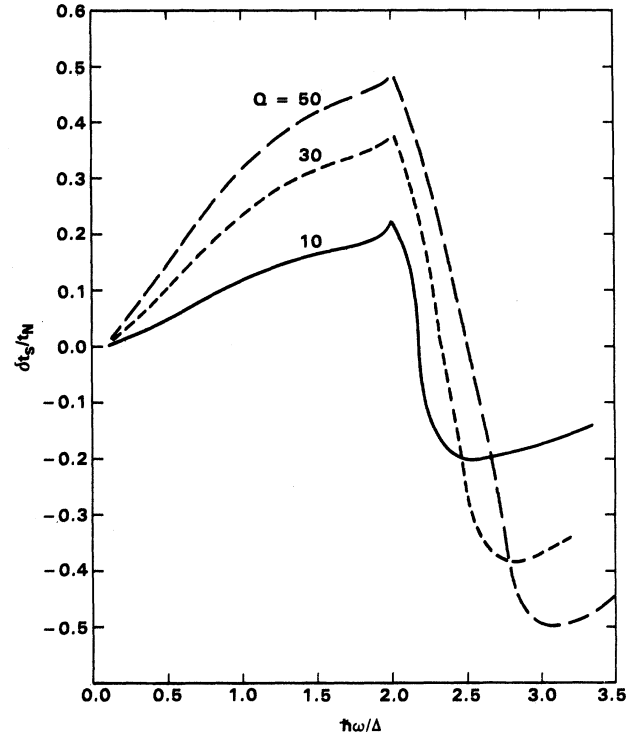


FIG. 5. The change in the transmittance  $\delta t_s$  of a superconducting thin film, between its equilibrium and externally driven steady-state, with respect to the normal-state transmittance  $t_N (= 0.337)$  vs the probe frequency, expressed as the photon energy divided by the nonequilibrium gap energy,  $\hbar\omega/\Delta$ . At an ambient temperature of  $T = 0.5T_c$ . For three different effective drive currents,  $Q = 10, 30,$  and  $50$  ( $Q' = 0.532, 1.597,$  and  $2.662$  for Al).

normalized frequency  $\mathcal{E}^*$ , for the case of a steady-state driving field with  $Q = 10, 30,$  and  $50$  when the ambient temperature is  $T/T_c = 0.5$  ( $Q' = 0.532, 1.597,$  and  $2.662$  for Al), and with  $R_N = 170 \Omega$  and  $n = 2.07$ . The solid lines in Fig. 6 show  $\delta t_s/t_N$  versus  $Q'$  for two fixed frequencies, for the same system as in Fig. 5. The solid lines in Fig. 7 are also  $\delta t_s/t_N$  versus  $Q'$ , but for the ambient temperature given by  $\beta = 7$  ( $T/T_c = 0.2518$ ).

For the same optical pulse as in Fig. 4, the time evolution of the thin-film transmission is depicted in Fig. 8. For Nb, the curves correspond to the frequencies 17, 60, and 135 GHz. The values of  $R_N$  and  $n$  (170  $\Omega$  and 2.07) are the same as in Fig. 5. The transient behavior of  $\delta t_s/t_s$  follows  $\delta n_{QP}/n_{QP}^0$ , reflecting the fact that, for these frequencies far below the pair-splitting frequency, the excess QP govern the fractional change in transmission. Figure 8(b) demonstrates that although the lowest frequencies exhibit the largest fractional change, it is the higher frequencies that experience the greatest absolute change. Within the pulse time  $t_0$ , at 135 GHz the transmittance increases from 0.033 to 0.08, whereas at 60 GHz it increases from 0.006 to 0.02.

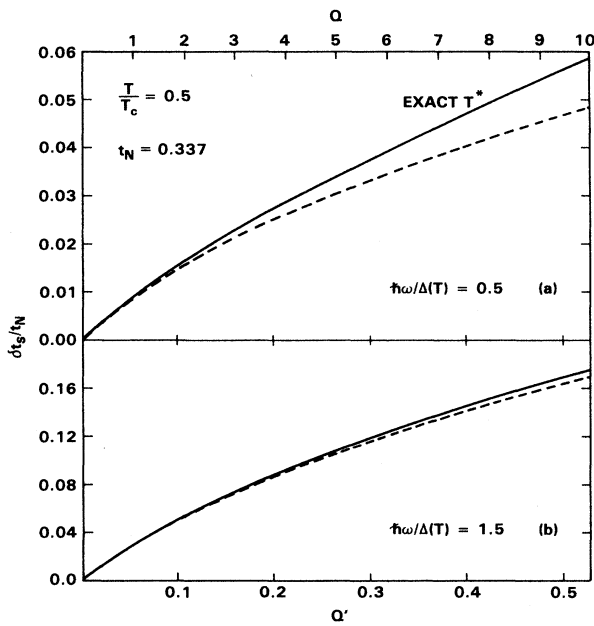


FIG. 6. The change in the transmittance  $\delta t_s$  of a superconducting thin film, between its equilibrium and externally driven steady state, with respect to the normal-state transmittance  $t_N$  ( $=0.337$  at low frequency) vs the effective external drive current:  $Q$  on the upper axis and temperature-independent  $Q'$  (for Al) on the lower axis. At ambient temperature  $T=0.5T_c$ . For two probe frequencies:  $\hbar\omega/\Delta(T)=0.5$  and  $1.5$ . Solid curves: the exact  $T^*$  model plus Mattis-Bardeen electrodynamics. Dashed curves: perturbative electrodynamics to first order in  $\delta\Delta/\Delta$ .

Laser radiation provides a convenient means to achieve optical control of microwave propagation in superconducting waveguides and strip lines. The attenuation and phase velocity in a superconducting waveguide are known functions of the conductivity (Refs. 15 and 16). We have used these functions together with the present theory of the laser-driven nonequilibrium state.<sup>17</sup> Figure 9 depicts the resulting frequency dependence of the equilibrium (solid curve) and laser-driven (dashed) steady-state attenuation and phase velocity of microwaves propagating in a Nb/Nb<sub>2</sub>O<sub>5</sub>/Nb waveguide. The Nb<sub>2</sub>O<sub>5</sub> is 0.1  $\mu\text{m}$  thick and the Nb films, which are equally illuminated, are several micrometers thick (the thick-film limit). The laser power, given by Eq. (2.53) with  $Q'=3$ , is 3–300 W/cm<sup>2</sup>, depending on  $\eta$ . Figure 9(a) reveals that laser radiation significantly enhances microwave attenuation below the pair-splitting frequency.<sup>18</sup> For example, at the interesting frequencies of 240, 135, and 94 GHz, the attenuation is increased from 34 to  $2 \times 10^4$  dB/m, from 22 to  $10^4$  dB/m, and 15 to  $6 \times 10^3$  dB/m, respectively. Thus, if a 2-mm strip is irradiated, it will, neglecting QP diffusion, decrease the intensity of a 240-GHz wave by 40 dB, a 135-GHz wave by 20 dB, and a 94-GHz wave by 12 dB.

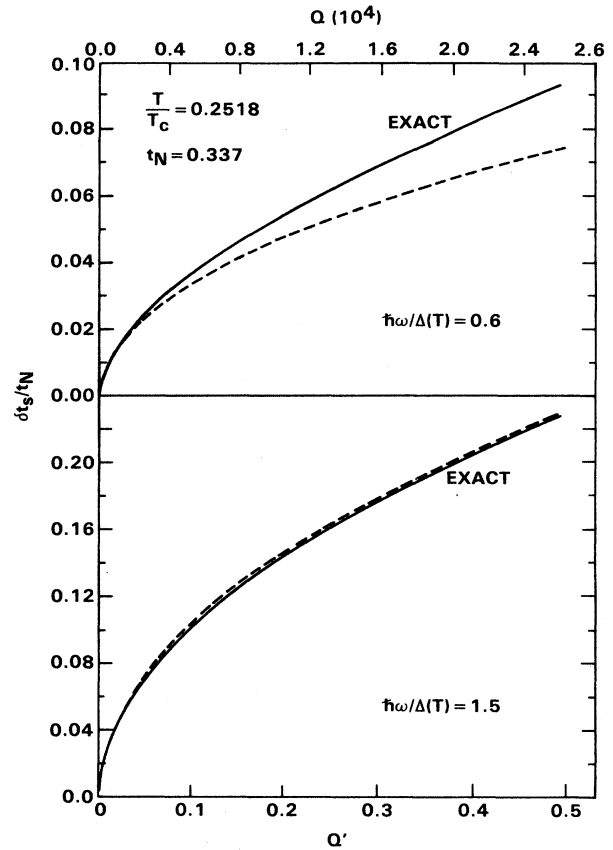


FIG. 7. Same as Fig. 6, except that the ambient temperature corresponds to  $\beta=7$ , or  $T \cong 0.2518T_c$ ; and the two probe frequencies are given by  $\hbar\omega/\Delta(T)=0.6$  and  $1.5$ .  $Q$  on the upper axis in units of  $10^4$ .

Figure 9(b) contrasts the phase velocity,  $V$ , in equilibrium with that under irradiation and reveals that below 710 GHz the equilibrium velocity exceeds the laser-driven one, whereas above 710 GHz the reverse is true. Below 100 GHz, the fractional change in phase velocity is about 10%, a useful feature for phase-shifting microwave radiation. Specifically, at 60 GHz the laser will decrease  $V$  from  $3.362 \times 10^7$  m/s to  $2.929 \times 10^7$  m/s, a change of nearly 15%. Accordingly, in the absence of QP diffusion, the difference in phase of a 60-GHz wave propagating over a path length of 1 mm in the presence versus the absence of laser radiation is  $94^\circ$ . Since the attenuation over 1 mm is 3 dB, such a system might be useful as a phase shifter. At 17 GHz, there is virtually no attenuation and the phase shift is about  $30^\circ$  over the same path length.

We have also studied the transient behavior of microwave propagation in this same Nb/Nb<sub>2</sub>O<sub>5</sub>/Nb waveguide when it is irradiated by  $1.2\bar{\tau}_R$  laser pulse. Figure 10 depicts the time evolution of the attenuation and phase velocity for 17-GHz and 240-GHz waves. The attenuation curve at 240-GHz is especially interesting for

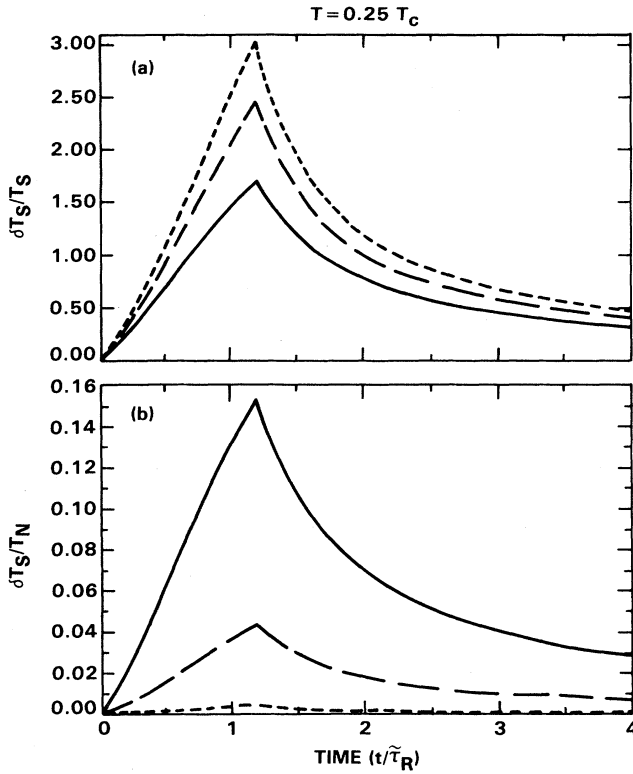


FIG. 8. Time evolution of change in microwave transmittance induced by light pulse of duration  $t_0 = 1.2\tau_R$  and strength  $Q = 1.6 \times 10^5$  at  $T = 0.25T_c$  ( $Q' = 3$  for Nb). Normalized in (a) to its equilibrium superconducting value and in (b) to its normal-state value ( $t_N = 0.037$ ). For three frequencies, which in Nb correspond to 135 GHz (solid line), 60 GHz (long-dashed), and 17 GHz (short-dashed line).

its applicability to millimeter-wave modulation. During the time  $t_0 = 1.2\tau_R$  (e.g., 1.2 ns for  $\eta = 0.73$ ) that the laser is on, the attenuation rises from 34 to  $1.46 \times 10^4$  dB/m—the peak attenuation across a 1-mm strip being 15 dB. Recovery of the equilibrium value of the attenuation is longer than the rise time; nevertheless, in just  $12\tau_R$  the attenuation falls to 1 dB over the 1-mm strip. Faster recovery times can be achieved by going to higher operating temperatures: raising  $T/T_c$  from 0.25 to 0.5 will cut the recovery time by a factor of 40 (i.e., to  $\tau_R/4$ ). The time evolution of the attenuation tracks that of the QP. On the other hand, the phase velocity recovers its equilibrium value much faster: After dropping from  $3.35 \times 10^7$  m/s to a minimum of  $3.07 \times 10^7$  m/s during the pulse time, the phase velocity recovers 85% of its lost velocity in the next  $4t_0$  (e.g., 4.7 ns for  $\eta = 0.73$ ).

### B. Perturbative Formulation

In the above procedure for determining the electro-dynamical response, we determine the QP density  $n_{QP}/n_{QP}^0$  from solutions to the RT equations; we put the

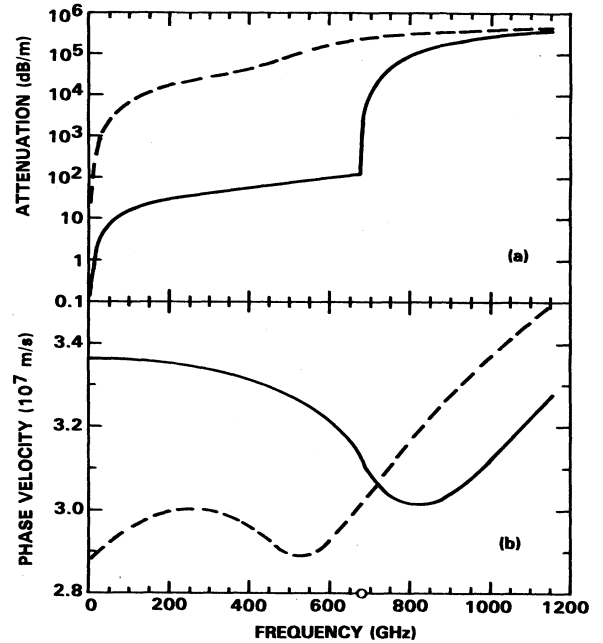


FIG. 9. Frequency dependence of the attenuation and phase velocity in a Nb/Nb<sub>2</sub>O<sub>5</sub>/Nb waveguide at  $T = 0.25T_c$ : (a) Attenuation in equilibrium (solid line) and in laser driven steady state (dashed line) with  $Q = 1.6 \times 10^5$ . (b) Phase velocity in equilibrium (solid line) and in laser-driven steady state (dashed line) with  $Q = 1.6 \times 10^5$  ( $Q' = 3$ ).

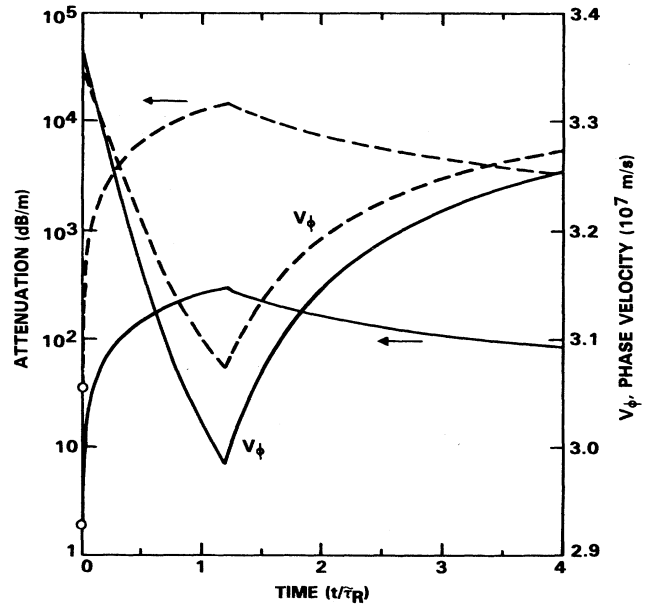


FIG. 10. Transient response of the attenuation and phase velocity of a Nb/Nb<sub>2</sub>O<sub>5</sub>/Nb waveguide at  $T = 0.25T_c$ . The response of 17 GHz (solid line) and 240 GHz (dashed line) guided waves to a laser pulse of duration  $1.2\tau_R$  with  $Q = 1.6 \times 10^5$  ( $Q' = 3$ ).

QP density into Eq. (3.8) and solve numerically for  $T^*$  [this requires numerically determining the value of the integral in Eq. (3.5) repeatedly in the equation solver]; and finally we use  $T^*$  in the numerical evaluation of the integrals in the MB expressions. When we solve for the case in which the fields vary in time, the passage from the solution  $n_{QP}(t)/n_{QP}^0$  to the time-dependent electrodynamic response, e.g.,  $\sigma(t)$ , requires numerically determining  $T^*$  and then the MB integrals at every "t" in some mesh. If we were to add spatial dependence as well, say with the solution  $n_{QP}(r,t)$  in Eq. (2.42), the entire procedure would have to be repeated over each point in the space mesh.

It would be convenient to have an equation directly relating  $\sigma$  to the solution  $n_{QP}(r,t)$ . We can develop such an expression by evaluating the MB equations to first order in  $\delta\Delta$  and  $\delta T$ , then relating  $\delta T$  to  $\delta\Delta$  through the BCS gap equation, and finally using the first-order approximation relating  $\delta\Delta$  to  $\delta n_{QP}$ . We now describe such a first-order perturbation theory and then show numerical comparisons between it and the exact  $T^*$  method—comparisons which demonstrate the method's usefulness.

Recalling that the complex conductivity from the MB equations has the functional form  $\sigma = \sigma(\mathcal{E} = \hbar\omega/\Delta, \beta = \Delta/kT)$  (where from here onward we shall understand that  $\sigma$  stands for the normalized conductivity  $\sigma/\sigma_n$ , unless the  $\sigma_n$  appears explicitly), we notice that the conductivity is a function of two variables whose values are changed by the external fields, namely,  $\Delta$  [where  $\delta\Delta = \Delta(T^*) - \Delta(T)$ ] and  $T$  [where  $\delta T = T^* - T$ ]. The BCS gap equation gives  $\Delta(T)$ , a function which can be inverted to yield  $\Delta^{-1} = T(\Delta)$ , so that  $\sigma$  is implicitly a function of  $\Delta$  alone. Thus the total derivative can be written as

$$\frac{d\sigma}{d\Delta} = \left[ \frac{\partial\sigma}{\partial\Delta} \right]_T + \left[ \frac{\partial\sigma}{\partial T} \right]_{\Delta} \frac{\partial T}{\partial\Delta}, \quad (4.4)$$

or

$$\begin{aligned} \delta\sigma &= \frac{d\sigma}{d\Delta} \delta\Delta = \left[ \frac{\partial\sigma}{\partial\Delta} \right]_T \delta\Delta + \left[ \frac{\partial\sigma}{\partial T} \right]_{\Delta} \frac{\partial T}{\partial\Delta} \delta\Delta \\ &= \left[ \frac{\partial\sigma}{\partial\Delta} \right]_T \delta\Delta + \left[ \frac{\partial\sigma}{\partial T} \right]_{\Delta} \delta T. \end{aligned} \quad (4.5)$$

Since  $\sigma$  depends explicitly on  $\Delta$  through  $\mathcal{E}$  and  $\beta$ , the partial derivatives become

$$\begin{aligned} \left[ \frac{\partial\sigma}{\partial\Delta} \right]_T &= \left[ \left[ \frac{\partial\sigma}{\partial\mathcal{E}} \right]_{\beta} \frac{\partial\mathcal{E}}{\partial\Delta} + \left[ \frac{\partial\sigma}{\partial\beta} \right]_{\mathcal{E}} \frac{\partial\beta}{\partial\Delta} \right]_T \\ &= \left[ - \left[ \frac{\partial\sigma}{\partial\mathcal{E}} \right]_{\beta} \mathcal{E} + \left[ \frac{\partial\sigma}{\partial\beta} \right]_{\mathcal{E}} \beta \right]_T \frac{1}{\Delta}, \end{aligned} \quad (4.6)$$

and

$$\left[ \frac{\partial\sigma}{\partial T} \right]_{\Delta} = \left[ \left[ \frac{\partial\sigma}{\partial\beta} \right]_{\mathcal{E}} \frac{\partial\beta}{\partial T} \right]_{\Delta} = - \frac{\beta}{T} \left[ \frac{\partial\sigma}{\partial\beta} \right]_{\mathcal{E}}. \quad (4.7)$$

Recalling the definition  $\mathcal{T} = T/T_c$  and introducing the

definition  $\Delta_N = \Delta/\Delta(0)$ , we notice that

$$\frac{\delta T}{T} = \frac{\partial T}{\partial\Delta} \frac{\delta\Delta}{T} = \frac{\partial\mathcal{T}}{\partial\Delta_N} \frac{T_c\Delta}{T\Delta(0)} \frac{\delta\Delta}{\Delta} = \frac{\partial\mathcal{T}}{\partial\Delta_N} \frac{\beta}{1.764} \frac{\delta\Delta}{\Delta}. \quad (4.8)$$

We now need to determine  $\partial\mathcal{T}/\partial\Delta_N$ . Again, using the weak-coupling approximation,  $\mathcal{T} = 1.764\Delta_N(1/\beta)$ , we have

$$\frac{\partial\mathcal{T}}{\partial\Delta_N} = 1.764 \left[ \frac{1}{\beta} + \Delta_N \frac{\partial}{\partial\Delta_N} \frac{1}{\beta} \right], \quad (4.9)$$

where the BCS gap equation tells us that  $\ln(\Delta_N) = F(\beta)$ , so that

$$\beta = F^{-1}(\ln(\Delta_N)), \quad (4.10)$$

and, here,  $F^{-1}$  is the inverse of the function defined by Eq. (3.7). After some algebra we find that

$$\frac{\partial}{\partial\Delta_N} \frac{1}{F^{-1}(\ln(\Delta_N))} = \frac{-1}{\Delta_N \beta^2 \frac{\partial F}{\partial\beta}}, \quad (4.11)$$

and thus

$$\frac{\partial\mathcal{T}}{\partial\Delta_N} = \frac{1.764}{\beta} \left[ 1 - \frac{1}{\beta} \frac{\partial F}{\partial\beta} \right], \quad (4.12)$$

where

$$\begin{aligned} \frac{\partial F}{\partial\beta} &= \frac{F}{\beta} + \frac{\beta}{2} \int_0^{\infty} x \sinh^{-1} x \operatorname{sech}^2 \left[ \frac{\beta}{2} (1+x^2)^{1/2} \right] \\ &\quad \times \tanh \left[ \frac{\beta}{2} (1+x^2)^{1/2} \right] dx. \end{aligned} \quad (4.13)$$

With this result, Eq. (4.8) becomes

$$\frac{\delta T}{T} = \left[ 1 - \frac{1}{\beta} \frac{\partial F}{\partial\beta} \right] \frac{\delta\Delta}{\Delta}. \quad (4.14)$$

Finally, Eqs. (4.6) and (4.7) substituted into Eq. (4.5), together with Eq. (4.14), gives us

$$\delta \left[ \frac{\sigma}{\sigma_n} \right] = H(\mathcal{E}, \beta) \frac{\delta\Delta}{\Delta} = [H_1(\mathcal{E}, \beta) + iH_2(\mathcal{E}, \beta)] \frac{\delta\Delta}{\Delta} \quad (4.15)$$

where

$$H(\mathcal{E}, \beta) = -\mathcal{E} \left[ \frac{\partial}{\partial\mathcal{E}} \frac{\sigma}{\sigma_n} \right]_{\beta} + \frac{1}{\partial F/\partial\beta} \left[ \frac{\partial}{\partial\beta} \frac{\sigma}{\sigma_n} \right]_{\mathcal{E}}. \quad (4.16)$$

To evaluate this expression for  $H$ , we must determine the derivatives, with respect to  $\mathcal{E}$  and  $\beta$ , of the four MB expressions for the real and imaginary parts of  $\sigma/\sigma_n$ , both

for  $\hbar\omega > 2\Delta$  and  $\hbar\omega < 2\Delta$ . The MB expressions for  $\sigma/\sigma_n$  involve integrals with square-root singularities. With some simple changes of variables, these singularities can be removed, the derivatives calculated, and the resulting integrals obtained numerically with rapid convergence. This process is outlined in the Appendix and the resulting expressions are given there.

$$\delta \left[ \frac{t_s}{t_N} \right] = -2(1-t_N^{1/2}) \left[ \frac{t_s}{t_N} \right]^2 \left\{ \left[ t_N^{1/2} + (1-t_N^{1/2}) \left[ \frac{\sigma_1}{\sigma_n} \right] \right] H_1 + (1-t_N^{1/2}) \left[ \frac{\sigma_2}{\sigma_n} \right] H_2 \right\} \frac{\delta\Delta}{\Delta}, \quad (4.17)$$

and that

$$\frac{\delta t_s}{t_s} = -2(B_1^2 + B_2^2)^{-1} (B_1 H_1 + B_2 H_2) \frac{\delta\Delta}{\Delta}, \quad (4.18)$$

where in this last equation we have introduced the definitions

$$B_1 = (t_N^{-1/2} - 1)^{-1} + \frac{\sigma_1}{\sigma_n} \quad (4.19a)$$

and

$$B_2 = \frac{\sigma_2}{\sigma_n}. \quad (4.19b)$$

These equations for the change in transmittance of a superconductor driven from equilibrium, with respect to its transmittance in either the normal or the equilibrium-superconducting state, are material dependent only through the value of  $t_N$  appearing on the right-hand sides.

Finally, we note that all of the above expressions for the changes in electrodynamical response functions are directly proportional to  $\delta\Delta/\Delta$ , which itself can be expressed in terms of the change in QP density, by use of the well-known first-order approximation to Eq. (3.1):

$$\frac{\Delta}{\Delta(0)} = 1 - 2n, \quad (4.20)$$

where  $n = n_{QP}/4N(0)\Delta(0)$  now refers to the nonequilibrium QP density. Equation (4.20) leads to

$$\frac{\delta\Delta}{\Delta(T)} = -\frac{\delta n_{QP}}{n_{QP}^0} \left[ \left[ \frac{\Delta(T)}{\Delta(0)} \right]^{-1} - 1 \right]. \quad (4.21)$$

As mentioned in Sec. II, the response of the QP density to the driving currents can be highly nonlinear for weak drive fields (e.g.,  $Q' \cong 10^{-4}$  at  $T=0.25T_c$ ). How good is the linear relation in Eq. (4.20) far from equilibrium (e.g.,  $Q' \cong 1$ , at  $T=0.25T_c$ )? Calculations of  $\delta\Delta/\Delta$  versus  $\delta n_{QP}/n_{QP}^0$ , with the exact  $T^*$  scheme and with Eq. (4.21), for  $T=0.25T_c$  and  $0.5T_c$  and out to  $Q' \cong 5$  (for Al), show a 3% deviation of the approximation from the exact result.

When Eq. (4.21) for  $\delta\Delta/\Delta$  is substituted into the equations for  $\delta\sigma$  or  $\delta t_s$ , we see the following: At a given reduced temperature  $\mathcal{T}=T/T_c$  (i.e., for a given  $\beta$ ) the value of  $\Delta(T)/\Delta(0)$ —needed in Eq. (4.21)—is deter-

The final result shown in Eq. (4.15) for the normalized change in conductivity is material independent: It is a universal property of BCS superconductors, in the same sense as the original MB expressions themselves.

Returning to the Glover-Tinkham expressions for the thin-film transmittance, as described in Sec. IV A, we find that

mined (by the BCS gap equation), and  $H(\mathcal{E},\beta)$  as a function of the normalized frequency ( $\mathcal{E}=\hbar\omega/\Delta$ ) is also determined. Thus, the complete time (and space) dependence of the nonequilibrium response functions  $[\sigma(\mathbf{r},t)]$  only requires our substituting the solution for  $n_{QP}(\mathbf{r},t)$  from the rate equations. The complex function  $H(\mathcal{E},\beta)$  is a universal function, in the sense that it applies to any BCS (weak-coupling) superconductor driven from equilibrium, independent of the material properties and independent of the strength of the driving force or of its space and time dependence. We only need to tabulate the values of  $H(\mathcal{E},\beta)$  once, for all nonequilibrium superconducting electrodynamic, regardless of the functional form of the driving field (subject, of course, to the aforementioned limitations of the theory itself).

Although we have developed this perturbation theory principally with the aim of facilitating calculations for arbitrarily complicated time (and/or space) dependence, it is most straightforward and reasonable to test it for the case of a uniform steady-state driving field. From Eq. (2.19), we know that

$$\delta n_{QP}/n_{QP}^0 = (n_{QP}^{ss} - n_{QP}^0)/n_{QP}^0 = (1+Q)^{1/2} - 1;$$

and, thus, at  $T/T_c=0.5$  we have that

$$\delta\Delta/\Delta = -0.045[(1+Q)^{1/2} - 1].$$

Using this result in Eq. (4.17), for example, we can find the change in transmittance,  $\delta t_s/t_N$ , which is shown in Fig. 6 (dashed curves) as a function of  $Q'$ . Figure 6 allows us to compare the perturbative results with those of the exact  $T^*$  model. The figure, which goes out to  $Q=10$  ( $Q' \cong 0.5$  for Al)—well beyond where the linearized kinetics breaks down—indicates that the perturbative theory, to first order in  $\delta\Delta/\Delta$ , gives a reasonable approximation (at  $Q=10$ , there is a 17% deviation for  $\mathcal{E}=0.5$  and only a 2.6% deviation for  $\mathcal{E}=1.5$ ).

Figure 7 presents the perturbative results (dashed curves) next to the exact  $T^*$  results for  $T/T_c=0.2518$ . Although the values of  $Q$  are 4 orders of magnitude greater in Fig. 7 than in Fig. 6, with  $\delta n/n$  thus 2 orders greater (for the same range of  $Q'$ , i.e., the same external drive strength), the approximation for  $\delta t_s/t_N$  is almost as good.

A conclusion of the numerical comparisons is that the conductivity change remains linear in the gap change well beyond where the gap change itself has departed

from linearity as a function of the driving current. It is very easy, especially at low temperatures ( $T \approx 0.25T_c$ ), to be in a regime where the QP density is a strongly nonlinear function of the external drive (laser intensity), but the gap change is linear in the QP density change, while the conductivity change is linear in the gap change. In other words, it is legitimate and useful to linearize the electrodynamics while by necessity retaining a nonlinear theory of the kinetics.

## V. DISCUSSION AND CONCLUSION

In this paper we have examined a formalism for treating superconductors that are driven from equilibrium by external fields—specifically optical laser fields—whose amplitudes may vary in time and/or space. This formalism is based on the framework of Parker's  $T^*$  model, incorporating the RT and MB equations. To solve the RT equations with time and/or space dependence, we have introduced an adiabatic elimination of the phonons from the kinetics. We have shown how elimination of the phonons from the RT equations leads to a single equation for the QP.

In applying this formalism, our interest focused specifically on the QP response to a pulsed optical laser field, and on the resulting, transient, microwave electrodynamics of superconducting thin films and thin-film waveguides. We presented analytic solutions for the QP response to a pulsed-laser field, of arbitrary strength, and to a weak oscillating electromagnetic grating. Our numerical implementation of the theory was performed for the case of superconducting Al and Nb films illuminated by a uniform pulsed laser.

The laser pulses considered here were of length  $1.2\bar{\tau}_R = (22 \text{ ps})(1 + \tau_{es}/\tau_B)$ , which for a reasonable value of around 50 for  $\tau_{es}/\tau_B$  means nanosecond time scales. Recovery times were seen to be around 10 times the rise time at  $0.25T_c$  and 40 times faster at  $0.5T_c$ . Within this time scale, at  $0.25T_c$ , a (60–200)-fold increase in the real part of the conductivity was calculated for frequencies of 17 to 240 GHz. An accompanying 4 to 2.5 fold increase in the thin-film transmittance was found, suggesting the possibility of an optically controlled switch for microwave radiation. Required power levels are from  $\text{mW}/\text{cm}^2$  (Al) to a few  $\text{W}/\text{cm}^2$  (Nb).

On the same time scale, the laser pulse was calculated to enhance the attenuation of Nb/Nb<sub>2</sub>O<sub>5</sub>/Nb waveguide

from 20 to  $2 \times 10^4$  dB/m, suggesting the possibilities of a microwave intensity modulator using pairs of appropriately timed laser pulses. Faster times might be achieved by using superconductors with faster recombination times—either by raising the reservoir temperature (thereby increasing the small microwave losses of the device) or by utilizing materials with a higher  $T_c$ , such as NbN or possibly the new ceramic oxides. Lower laser powers can be achieved with (1) a light source that optimizes the ratio of absorptance to penetration depth, (2) a substrate that increases the value of  $\tau_{es}/\tau_B$  [though at the sacrifice of slower speeds—see Eq. (2.53)], or (3) thinner films.

Still on the same time scales, but for the longer wavelengths ( $\leq 60$  GHz), the calculations here showed that an optically driven Nb/Nb<sub>2</sub>O<sub>5</sub>/Nb waveguide can significantly depress the phase velocity without serious attenuation: Almost a 90° phase shift was calculated for a 60-GHz wave over a propagation distance of 1 mm. Since it is usually sufficient to operate on a microsecond scale for phase shifting applications, larger values of  $\tau_{es}/\tau_B$  could be used, to permit very low laser intensities to achieve large phase shifts.

In order to treat the problem of QP diffusion in the laser field, we shall have to solve the QP kinetic equation, Eq. (2.17), numerically. Diffusion codes exist for such purposes. To go from the solution for the QP density to the electrodynamics (assuming always that the conditions for the validity of the MB equations hold) may be very computer intensive. It would be very convenient to have a direct relationship between the change in the conductivity and the change in the QP density. Such a relationship would also link the conductivity change to the fundamental superconducting time and distance scales, and would greatly facilitate extracting information on the properties of superconducting thin films under nonequilibrium conditions from measurement of the perturbed microwave characteristics. For example, if the drive currents are pulsed, one can directly extract the nonequilibrium QP recombination time by measuring the time evolution of the change in the microwave transmission,  $\delta t_s$ , as discussed in Sec. IV. To that end, we have presented here a perturbative theory, based on a series expansion of the MB equations to first order. Our numerical work has shown that such equations for the electrodynamic response carried out to first order in  $\delta n_{QP}/n_{QP}^0$ , are valid for  $Q'$  on the order of unity—a value for which the QP response, at even moderately low temperatures, can be deep within the nonlinear regime.

## APPENDIX

We present here the expressions for the derivatives that appear in Eq. (4.16) defining the function  $H(\mathcal{E}, \beta)$ . From the Mattis-Bardeen equations, with the definitions that  $\beta = \Delta/kT$  and  $\mathcal{E} = \hbar\omega/\Delta$  [where  $\Delta = \Delta(T)$  at equilibrium], we see that for  $\hbar\omega < 2\Delta$

$$\frac{\sigma_1}{\sigma_n} \equiv \frac{\sigma_1^<}{\sigma_n} = \frac{2}{\mathcal{E}} \int_1^\infty [\tilde{f}(E) - \tilde{f}(E + \mathcal{E})] g(E) dE, \quad (\text{A1})$$

where

$$\tilde{f}(E) = \frac{1}{\exp(\beta E) + 1} \tag{A2}$$

and

$$\tilde{g}(E) = \frac{1 + E^2 + \mathcal{E}E}{(E^2 - 1)^{1/2} [(E + \mathcal{E})^2 - 1]^{1/2}} . \tag{A3}$$

Then after we make the substitution  $E = \cosh x$ , we find that

$$\frac{\partial}{\partial \beta} \left[ \frac{\sigma_1}{\sigma_n} \right]_{\mathcal{E}} = \frac{-2}{\mathcal{E}} \int_0^\infty [f_a^2(x) \cosh x - f_b^2(x) e^{\beta \mathcal{E}} (\cosh x + \mathcal{E})] e^{\beta \cosh x} g_a(x, \mathcal{E}) dx , \tag{A4}$$

where

$$f_a(x) = \tilde{f}(\cosh x), \quad f_b(x) = \tilde{f}(\cosh x + \mathcal{E}), \quad \text{and} \quad g_a(x, \mathcal{E}) = \tilde{g}(\cosh x) . \tag{A5}$$

Similarly, we have that

$$-\mathcal{E} \frac{\partial}{\partial \mathcal{E}} \left[ \frac{\sigma_1}{\sigma_n} \right]_{\beta} = 2 \int_0^\infty [f_a - f_b] \left[ \frac{g_a}{\mathcal{E}} - \frac{\partial g_a}{\partial \mathcal{E}} \right] dx - 2\beta \int_0^\infty f_b^2 e^{\beta(\mathcal{E} + \cosh x)} g_a dx , \tag{A6}$$

where

$$\frac{\partial g_a(x, \mathcal{E})}{\partial \mathcal{E}} = \frac{-(2 \cosh x + \mathcal{E})}{[(\cosh x + \mathcal{E})^2 - 1]^{3/2}} . \tag{A7}$$

Continuing with the case of  $\hbar\omega < 2\Delta$ , we write the MB equation for the imaginary part as

$$\begin{aligned} \frac{\sigma_2}{\sigma_n} &= \frac{1}{\mathcal{E}} \int_{1-\mathcal{E}}^1 [1 - 2\tilde{f}(E + \mathcal{E})] q(E) dE \\ &= \frac{1}{\mathcal{E}} \left[ \int_{1-\mathcal{E}}^{1-\mathcal{E}/2} + \int_{1-\mathcal{E}/2}^1 \right] [1 - 2\tilde{f}(E + \mathcal{E})] q(E) dE , \end{aligned} \tag{A8}$$

where

$$q(E) = \frac{1 + E(E + \mathcal{E})}{(1 - E^2)^{1/2} [(E + \mathcal{E})^2 - 1]^{1/2}} . \tag{A9}$$

We have broken the integral into two pieces; for the first we now use the substitution  $E + \mathcal{E} = \cosh x$ , and for the second we use  $E = \sin x$ . Then after some algebra, we obtain the following results:

$$\frac{\partial}{\partial \beta} \left[ \frac{\sigma_2}{\sigma_n} \right]_{\mathcal{E}} = \frac{2}{\mathcal{E}} \int_0^{\cosh^{-1}(1+\mathcal{E}/2)} f_a^2(x) e^{\beta \cosh x} (\cosh x) C(x) dx + \frac{2}{\mathcal{E}} \int_{\sin^{-1}(1-\mathcal{E}/2)}^{\pi/2} f_c^2(x) e^{\beta(\sin x + \mathcal{E})} (\sin x + \mathcal{E}) S(x) dx \tag{A10}$$

and

$$\begin{aligned} -\mathcal{E} \frac{\partial}{\partial \mathcal{E}} \left[ \frac{\sigma_2}{\sigma_n} \right]_{\beta} &= -[1 - 2\tilde{f}(1 + \mathcal{E}/2)] \frac{2 - (\mathcal{E}/2)^2}{\frac{\mathcal{E}}{2} [4 - (\mathcal{E}/2)^2]^{1/2}} + \int_{\sin^{-1}(1-\mathcal{E}/2)}^{\pi/2} (1 - 2f_c) \left[ \frac{S}{\mathcal{E}} - \frac{\partial S}{\partial \mathcal{E}} \right] dx \\ &\quad + \int_0^{\cosh^{-1}(1+\mathcal{E}/2)} (1 - 2f_a) \left[ \frac{C}{\mathcal{E}} - \frac{\partial C}{\partial \mathcal{E}} \right] dx - 2\beta \int_{\sin^{-1}(1-\mathcal{E}/2)}^{\pi/2} f_c^2 e^{\beta(\sin x + \mathcal{E})} S dx , \end{aligned} \tag{A11}$$

where the following definitions have been used:

$$f_c(x) = \tilde{f}(\sin x + \mathcal{E}) , \tag{A12}$$

$$C(x) = \frac{1 + \cosh x (\cosh x - \mathcal{E})}{[1 - (\cosh x - \mathcal{E})^2]^{1/2}} , \tag{A13}$$

$$S(x) = \frac{1 + \sin x (\sin x + \mathcal{E})}{[(\sin x + \mathcal{E})^2 - 1]^{1/2}} , \tag{A14}$$

and thus,

$$\frac{\partial C}{\partial \mathcal{E}} = \frac{-2 \cosh x + \mathcal{E}}{[1 - (\cosh x - \mathcal{E})^2]^{3/2}} \quad \text{and} \quad \frac{\partial S}{\partial \mathcal{E}} = \frac{-(2 \sin x + \mathcal{E})}{[(\sin x + \mathcal{E})^2 - 1]^{3/2}} . \tag{A15}$$



We now turn to the case in which  $\hbar\omega > 2\Delta$ . First, for the real part, we have

$$\begin{aligned} \frac{\sigma_1}{\sigma_n} &\equiv \frac{\sigma_1^>}{\sigma_n} = \frac{\sigma_1^<}{\sigma_n} + \frac{1}{\mathcal{E}} \int_{(1-\mathcal{E})}^{-1} [1 - 2\tilde{f}(E + \mathcal{E})] |\tilde{g}(E)| dE \\ &= \frac{\sigma_1^<}{\sigma_n} - \frac{1}{\mathcal{E}} \left[ \int_{(1-\mathcal{E})}^{-\mathcal{E}/2} + \int_{-\mathcal{E}/2}^{-1} \right] [1 - 2\tilde{f}(E + \mathcal{E})] \tilde{g}(E) dE . \end{aligned} \quad (\text{A16})$$

In the above equation, the first term,  $\sigma_1^</math>/ $\sigma_n$ , refers to the expression in Eq. (A1), and the integral has been broken into two parts. For the first integral we make the substitution  $E + \mathcal{E} = \cosh x$  and for the second we use  $E = -\cosh x$ , and then after performing the differentiation, we arrive at the following results:$

$$\begin{aligned} \frac{\partial}{\partial \beta} \left[ \frac{\sigma_1}{\sigma_n} \right]_{\mathcal{E}} &\equiv \frac{\partial}{\partial \beta} \left[ \frac{\sigma_1^>}{\sigma_n} \right]_{\mathcal{E}} = \frac{\partial}{\partial \beta} \left[ \frac{\sigma_1^<}{\sigma_n} \right]_{\mathcal{E}} \\ &\quad - \frac{2}{\mathcal{E}} \int_0^{\cosh^{-1}(\mathcal{E}/2)} [f_a^2(x) \cosh x e^{\beta \cosh x} + f_d^2(x) (\mathcal{E} - \cosh x) e^{\beta(\mathcal{E} - \cosh x)}] Q(x, \mathcal{E}) dx , \end{aligned} \quad (\text{A17})$$

where the first term on the right-hand side,  $\partial(\sigma_1^</math>/ $\sigma_n)/\partial\beta$ , is given by the expression in Eq. (A4),$

$$f_d(x) = \tilde{f}(\mathcal{E} - \cosh x) \quad \text{and} \quad Q(x, \mathcal{E}) = g_a(x, -\mathcal{E}) , \quad (\text{A18})$$

and

$$\begin{aligned} -\mathcal{E} \frac{\partial}{\partial \mathcal{E}} \left[ \frac{\sigma_1}{\sigma_n} \right]_{\beta} &\equiv -\mathcal{E} \frac{\partial}{\partial \mathcal{E}} \left[ \frac{\sigma_1^>}{\sigma_n} \right]_{\beta} = -\mathcal{E} \frac{\partial}{\partial \mathcal{E}} \left[ \frac{\sigma_1^<}{\sigma_n} \right]_{\beta} - 1 \\ &\quad + 2\tilde{f}(\mathcal{E}/2) - 2 \int_0^{\cosh^{-1}(\mathcal{E}/2)} [1 - f_a(x) - f_d(x)] \left[ \frac{Q}{\mathcal{E}} - \frac{\partial Q}{\partial \mathcal{E}} \right] dx \\ &\quad - 2\beta \int_0^{\infty} f_b^2(x) e^{\beta(\mathcal{E} + \cosh x)} g_a(x, \mathcal{E}) dx \\ &\quad + 2\beta \int_0^{\cosh^{-1}(\mathcal{E}/2)} f_d^2(x) e^{\beta(\mathcal{E} - \cosh x)} Q(x, \mathcal{E}) dx , \end{aligned} \quad (\text{A19})$$

where

$$\frac{\partial Q}{\partial \mathcal{E}} = \frac{2 \cosh x - \mathcal{E}}{[(\cosh x - \mathcal{E})^2 - 1]^{3/2}} . \quad (\text{A20})$$

Next, for the imaginary part, and  $\hbar\omega > 2\Delta$ , we have

$$\frac{\sigma_2}{\sigma_n} = \frac{1}{\mathcal{E}} \int_{-1}^1 [1 - 2\tilde{f}(E + \mathcal{E})] q(E) dE . \quad (\text{A21})$$

With the substitution  $E = \sin x$ , we eventually find

$$\frac{\partial}{\partial \beta} \left[ \frac{\sigma_2}{\sigma_n} \right]_{\mathcal{E}} = \frac{2}{\mathcal{E}} \int_{-\pi/2}^{\pi/2} f_c^2(x) e^{\beta(\sin x + \mathcal{E})} (\sin x + \mathcal{E}) S(x) dx \quad (\text{A22})$$

and

$$-\mathcal{E} \frac{\partial}{\partial \mathcal{E}} \left[ \frac{\sigma_2}{\sigma_n} \right]_{\beta} = \int_{-\pi/2}^{\pi/2} [1 - 2f_c(x)] \left[ \frac{S}{\mathcal{E}} - \frac{\partial S}{\partial \mathcal{E}} \right] dx - 2\beta \int_{-\pi/2}^{\pi/2} f_c^2(x) e^{\beta(\sin x + \mathcal{E})} S(x) dx . \quad (\text{A23})$$

<sup>1</sup>Nonequilibrium Superconductivity, edited by D. N. Langenberg and A. I. Larkin (North-Holland, New York, 1986).

<sup>2</sup>Nonequilibrium Superconductivity, Phonons, and Kapitza Boundaries, Vol. 65 of NATO Advanced Study Institute, Series B, edited by Kenneth G. Gray (Plenum, New York, 1981).

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289–340 in Ref. 2, or the model by I. Iguchi and H. Konno, Phys. Rev. B **28**, 4040 (1983).

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- <sup>18</sup>The pair-splitting frequency in Fig. 9 is 8% below the measured value, because we used the ideal BCS weak-coupling approximation  $\Delta(0) = 1.764kT_c$  with the experimental value of  $T_c$ .