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Third-harmonic generation in dirty superconductors

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The third-harmonic response of a superconductor near its transition temperature to a weak electromagnetic field is calculated, both in the gap and gapless regimes, including the effect of inelastic scattering of electrons by the phonons. The response is related to the relaxation time of the order parameter in the perturbed superconductor; it is shown that the temperature dependence of the relaxation time, depending on the depairing strength, is changed as T_c is approached from below.

I. INTRODUCTION

In a recent article, Amato and McLean[1] reported a measurement of the order-parameter relaxation time in a superconductor close to T_c. The relaxation time describes the return to equilibrium of a superconductor disturbed by a fairly high-frequency field ($\omega = 11$ GHz), and is measured by observing the third-harmonic response of the superconductor. The theoretical basis for the experiment was developed by Gorkov and Eliashberg.[2] However, that theory was designed for gapless superconductors, while the experiment was carried on a fairly dilute alloy. Since expressions for the third-harmonic response in this limit were not available, the authors of Ref. 1 analyzed their data on basis of the Gorkov-Eliashberg[2] theory, which was modified to include some results of Eliashberg,[3] developed for the case of extremely dilute alloys. With this modification, it was found that the temperature dependence of the relaxation time is $(T_c - T)^{-1}$ as T approaches T_c.

Here we derive expressions for the third-harmonic response, which in the limits of high and low concentrations of magnetic impurities, respectively, yield the results of the Gorkov-Eliashberg and Eliashberg theories. We therefore believe that the expressions presented here fill the gap which has existed so far. We also include in the calculation the effect of inelastic scattering of the electrons by the phonons. The following results are obtained: For gapless superconductors we recover the Gorkov-Eliashberg[2] expression, in which the relaxation time behaves as $(T_c - T)^{-1}$. In the case of dilute alloys ($\rho \ll 1$, where ρ is the pair-breaking parameter) we distinguish between two cases: the gapless regime, which occurs close to T_c, and the gap regime, dominating when the order parameter Δ is such that $\Delta > \rho 2\pi T$. In the first case we find the $(T_c - T)^{-1}$ behavior, while in the second the temperature dependence of the relaxation time is $(T_c - T)^{-3/2}$. We also find that when the electronic life-time τ_E due to inelastic collisions with the phonons is such that $1 \ll \Delta^2 \tau_E / T \ll \kappa^2$, where κ is the Landau-Ginzburg parameter, the relaxation time in the gap regime behaves like $\tau_E [(T_c - T)/T]^{-1/2}$ (for $\Delta / T \gg \rho$).

The outline of the paper is as follows. In Sec. II we exhibit the equations describing the nonstationary behavior of the order parameter, disturbed from equilibrium by a frequency-dependent field. Naturally, these equations include the electromagnetic field. We therefore add to them an equation resulting from the Maxwell equations, which gives the field behavior inside the superconductor. This closes the set of equations needed for the solution of the problem at hand. In the equation for the field appears a term proportional to

$$\Delta^2(Z,t) A(Z,t), \tag{1}$$

in which A is the vector potential and $\Delta(Z,t) = \Delta + \delta\Delta_1(Z,t)$, where Δ is the equilibrium value of the order parameter. (The geometry of the problem is described in Sec. II.) In the equations for the variation of $\delta\Delta_1$ appears A^2. We insert for it the zero-order value of the field, namely, treat the term in equation (1) as $\Delta^2 A(Z,t)$. We therefore find that $\delta\Delta_1(Z,t)$ is proportional to $A^2 \sim e^{-2i\omega t}$. This solution is inserted into the equation for A, and as a result there appears a term $e^{-3i\omega t}$ in the first-order correction to A. [See Eq. (1)]. This is the origin of the third-harmonic response. In Sec. III we analyze the frequency regime in which $\delta\Delta_1(Z,t)$ follows the spatial variation of the field and calculate the third-harmonic amplitude in the cases mentioned above. From this, the relaxation time of the order parameter is found.

II. GENERAL EQUATIONS

We consider a superconductor occupying the half-space $Z < 0$, on which a weak high-frequency magnetic field is applied parallel to the surface. Consequently, the superconducting order parameter is disturbed from its equilibrium value. Our aim is to study its response to the field at the vicinity of the transition temperature.

The general equations describing nonstationary properties of a superconductor close to T_c were first derived by Gorkov and Eliashberg[2] and Eliashberg.[3,4] They were rederived and analyzed by Schmid and Schon[5] and us.[6] (See specifically Appendixes A–C in Ref. 6.) It turns out that the equations for the order parameter, as well as those for the charge and current densities, consist of two parts. The first is the regular part, which, as the external frequency tends to zero, goes over to the corresponding static expression; e.g., the static Ginzburg-Landau equation. The second is called the anomalous part and appears only below T_c. Its origin is in the deviation from equilibrium of the quasiparticle distribution function, and hence it is sensitive to the details of the energy spectrum.

In the present problem, $\Delta(Z,t)$ can be chosen to be real.[3] The nonstationary Ginzburg-Landau equation for $\Delta(Z,t)$ in the dirty limit, is (see Refs. 2–6),

$$\left[\frac{\psi'(\frac{1}{2}+\rho)}{4\pi T}\left(-\frac{\partial}{\partial t}+D\frac{\partial^2}{\partial Z^2}-\frac{2e^2}{c}DA^2(Z,t)\right)+\alpha_0-\beta_0\Delta^2(Z,t)\right]\Delta(Z,t)+\Psi(Z,t)=0. \tag{2}$$

(We use dimensions in which $\hbar=k_B=1$.) The following notation has been introduced in (2): The depairing parameter is ρ, and it enters in the polygamma functions $\psi^{(n)}(x)$. It contains the conduction electron spin-flip time τ_s due to collision with magnetic impurities, and the time τ_E due to inelastic scattering with the phonons

$$\rho=(1/2\pi T)(1/\tau_s+1/2\tau_E). \tag{3}$$

The diffusion coefficient is $D=\frac{1}{3}v_F^2\tau_1$, where v_F is the velocity at Fermi surface and τ_1 is the potential scattering time. In the dirty limit, $T\tau_1\ll 1$. The vector potential is A and

$$\alpha_0=[(T_c-T)/T_c][1-\rho\psi'(\tfrac{1}{2}+\rho)], \tag{4a}$$

$$\beta_0=-[1/(4\pi T)^2][\psi''(\tfrac{1}{2}+\rho)+\tfrac{1}{3}\rho\psi'''(\tfrac{1}{2}+\rho)]. \tag{4b}$$

Note that β_0 is positive and that at equilibrium $\Delta^2=\alpha_0/\beta_0$.

The last term on the left-hand side of (2) is the anomalous term. Explicitly, (see Refs. 5 and 6), it is given by a certain energy integral of the deviation of the quasiparticle distribution from equilibrium. This deviation obeys a Boltzman-type equation, which includes a collision integral due to inelastic scattering by the phonons. Here we treat it in a single relaxation-time approximation, namely, the collision integral is replaced by $1/\tau_E$. Consequently, one obtains

$$\left(\frac{\partial}{\partial t}-D\frac{\partial^2}{\partial Z^2}+\frac{1}{\tau_E}\right)\Psi(Z,t)=-I\frac{\partial}{\partial t}\Delta^2(Z,t), \tag{5}$$

where $I=I(\Delta,T,\rho)$. This quantity is calculated in the Appendix. The results depend on whether there is a gap in the excitation spectrum or the gap is smeared out by the pair breakers.

To close the set of Eqs. (2) and (5), we need an equation describing the variation of $A(Z,t)$ inside the superconductor. This will be derived now.

In the case of a neutral superconductor (in which the charge density vanishes), the current density is given by (see Refs. 2–6).

$$\vec{j}=\sigma\vec{E}-\frac{\psi'(\frac{1}{2}+\rho)}{2\pi T}\frac{2\sigma}{c}\Delta^2(Z,t)\vec{A}. \tag{6}$$

Here $\vec{E}$ is the electric field, $\sigma=Ne^2\tau_1/m=2De^2N(0)$, where N is the electron concentration, and $N(0)$ is the density of states at Fermi surface. Now, from the Maxwell equations, $\vec{j}=(c/4\pi)\vec{\nabla}\times\vec{H}$, where $\vec{H}$ is the magnetic field. In the present problem, in which the superconductor occupies the half space $Z<0$ and the magnetic field is parallel to it, the vector potential can be chosen to be parallel to the interface too. Moreover, it depends only on the coordinate Z. Hence, in the gauge in which the scalar potential vanishes, Eq. (6) yields

$$\frac{\partial^2}{\partial Z^2}A(Z,t)=\frac{4\pi\sigma}{c^2}\left(\frac{\partial A(Z,t)}{\partial t}+\frac{\psi'(\frac{1}{2}+\rho)}{\pi T}\Delta^2(Z,t)A(Z,t)\right). \tag{7}$$

Here the relation $E=-(1/c)(\partial A/\partial t)$ was used. The last term on the right-hand side is the origin of the third harmonic [see the discussion after Eq. (1)].

From (7) it is seen that the zero-order value of A [i.e., when in the last term $\Delta^2(Z,t)$ is replaced by the equilibrium value Δ^2], decays into the superconductor according to the law[2]

$$A(Z,t)=A_0e^{kZ-i\omega t}, \tag{8a}$$

$$k^2(\omega)=-\frac{4\pi\sigma}{c^2}i\omega+\frac{4\pi\sigma}{c^2}\frac{\psi'(\frac{1}{2}+\rho)}{\pi T}\Delta^2=-2i\delta_S^{-2}+\delta_L^{-2}. \tag{8b}$$

Here δ_S is the normal skin depth for a metal in the normal state and δ_L is the London penetration depth. At low frequencies the penetration of the field is determined by δ_L, while at high frequencies, the normal skin depth pertains. The frequency which separates the two regions and determines the low-frequency behavior of the field is ω_0,

$$\omega_0=[\psi'(\tfrac{1}{2}+\rho)/\pi T]\Delta^2. \tag{9}$$

For gapless superconductors, $\rho \gg 1$, and

$$\omega_0 \simeq \Delta^2/\pi T\rho \simeq 2\Delta^2\tau_S , \quad (10a)$$

which is the Gorkov-Eliashberg[2] result. (Note that in Ref. 2, τ_E was omitted.) For $\rho \ll 1$, we have

$$\omega_0 \simeq (\pi\Delta^2/2T)\,[1+(2\rho/\pi^2)\psi''(\tfrac{1}{2})\,] . \quad (10b)$$

Since $\psi''(\frac{1}{2})$ is negative, a small amount of pair breakers reduces ω_0 compared to its value for $\rho=0$. In any case ω_0 is given by $N_S/N\tau_1$, where N_S is the "super-electron" concentration.

We now insert the zero-order value of A [Eq. (8)] into Eqs. (2) and (5), to solve $\Delta(Z,t)$. Denoting $\Delta(Z,t)=\Delta+\delta\Delta_1(Z,t)$, and linearizing these equations, we obtain

$$\frac{\psi'(\frac{1}{2}+\rho)}{4\pi T}\left[\left(-\frac{\partial}{\partial t}+D\frac{\partial^2}{\partial Z^2}\right)\delta\Delta_1-\left(\frac{2e}{c}\right)^2 DA^2\Delta\right] - 2\beta_0\Delta^2\delta\Delta_1+\Psi=0, \quad (11a)$$

$$\left(\frac{\partial}{\partial t}-D\frac{\partial^2}{\partial Z^2}+\frac{1}{\tau_E}\right)\psi=-2I\Delta\frac{\partial}{\partial t}\delta\Delta_1 . \quad (11b)$$

The solution of Eq. (11) must obey the boundary conditions[2,3] at $Z=0$, namely, $(\partial\delta\Delta_1/\partial Z)_{Z=0}=0$, $(\partial\Psi/\partial Z)_{Z=0}=0$. After a straightforward, although cumbersome, calculation, one obtains the following solution:

$$\delta\Delta_1(Z,t)=e^{-2i\omega t}(ae^{2kZ}+a_1e^{k_1Z}+a_2e^{k_2Z}) , \quad (12)$$

in which k is given by (8b) and k_1, k_2 depend on ω. They are given by

$$\left(i\gamma\frac{\omega}{\omega_0}+\frac{(k_n\delta_L)^2}{2\kappa^2}-1\right)\times\left(i\gamma\frac{\omega}{\omega_0}+\frac{(k_n\delta_L)^2}{2\kappa^2}-\frac{\gamma}{2\omega_0\tau_E}\right)-i\gamma\frac{\omega}{\omega_0}\frac{\nu}{2}=0, \quad n=1,2 , \quad (13)$$

where ν, κ^2, and γ are dimensionless quantities:

$$\nu=2I/\beta_0\Delta , \quad (14a)$$

$$\gamma=(1/\beta_0)\,[\psi'(\tfrac{1}{2}+\rho)/2\pi T\,]^2 , \quad (14b)$$

$$\kappa^2=\omega_0\delta_L^2/D\gamma. \quad (14c)$$

The last quantity κ is the Landau-Ginzburg parameter. The coefficients a, a_1, and a_2 are given by

$$a=\tfrac{1}{4}\Delta\left(\frac{A_0}{\delta_L H_c}\right)^2\left(i\gamma\frac{\omega}{\omega_0}+\frac{(2k\delta_L)^2}{2\kappa^2}-\frac{\gamma}{2\omega_0\tau_E}\right)\Big/M , \quad (15a)$$

$$M=\left(i\gamma\frac{\omega}{\omega_0}+\frac{(2k\delta_L)^2}{2\kappa^2}-1\right)\times\left(i\gamma\frac{\omega}{\omega_0}+\frac{(2k\delta_L)^2}{2\kappa^2}-\frac{\gamma}{2\omega_0\tau_E}\right)-i\gamma\frac{\omega}{\omega_0}\frac{\nu}{2}, \quad (15b)$$

and

$$2k\,a+k_1a_1+k_2a_2=0, \quad (16a)$$

$$-k\,\Delta\left(\frac{\kappa A_0}{H_c\delta_L}\right)^2+(2k\,\delta_L)^2 2ka=-\delta_L^2(k_1^3a_1+k_2^3a_2). \quad (16b)$$

Here H_c is the temperature-dependent thermodynamic critical field.

The expression (12) for $\delta\Delta_1(Z,t)$ is now inserted into Eq. (7), for the field, and a first-order correction to A is found. This is done by linearizing the last term on the right-hand side of (7), $\Delta^2(Z,t)A \simeq \Delta^2A+2\Delta A\,\delta\Delta_1(Z,t)$. Using Eqs. (8) and (12), the first-order correction to the vector potential is

$$A_1(Z,t)=e^{-3i\omega t}(a_3e^{k(3\omega)Z}+a_4e^{3k(\omega)Z}+a_5e^{[k(\omega)+k_1(\omega)]Z}+a_6e^{[k(\omega)+k_2(\omega)]Z}). \quad (17)$$

The coefficients a_4, a_5, and a_6 are given in terms of the coefficients a, a_1, and a_2,

$$\{[3k(\omega)]^2-k^2(3\omega)\}a_4=(2A_0/\Delta\delta_L^2)a , \quad (18a)$$

$$\{[k(\omega)+k_1(\omega)]^2-k^2(3\omega)\}a_5=(2A_0/\Delta\delta_L^2)a_1 , \quad (18b)$$

$$\{[k(\omega)+k_2(\omega)]^2-k^2(3\omega)\}a_6=(2A_0/\Delta\delta_L^2)a_2. \quad (18c)$$

The coefficient a_3 cannot be found from (7). It is determined by the boundary conditions on the field. Now consider the case[2] in which the electric field $E_1=-(1/c)(\partial/\partial t)A_1$ vanishes at the surface $Z=0$. We then find that the third-harmonic component of the magnetic field at $Z=0$ is

$$H_1(Z=0,t)=(2A_0/\Delta\delta_L^2)e^{-3i\omega t}\times\left[a\frac{1}{3k(\omega)+k(3\omega)}+a_1\frac{1}{k(\omega)+k_1(\omega)+k(3\omega)}+a_2\frac{1}{k(\omega)+k_2(\omega)+k(3\omega)}\right] , \quad (19)$$

where (18) has been used. The amplitude of $H_1(Z=0,t)$ is the quantity measured in the experiment.[1] In Sec. III we study it in detail.

III. THIRD-HARMONIC RESPONSE

Here we investigate the frequency regimes in which the third-harmonic measurement can be used to study the relaxation time of the order parameter. We are interested in the case where the field penetration is determined by the Meissner effect, i.e., when [from Eqs. (8) and (9)]

$$\omega<\omega_0 . \quad (20)$$

Next we turn to study the spatial behavior of the order parameter, given in Eqs. (12)–(16). From Eq. (12) it is seen that $\delta\Delta_1$ will follow the spatial variation of the field when the last two terms on the right-hand side of (12) are negligible compared to the first term. To find the frequency regime in which this behavior occurs, we proceed as follows. Equation (13) yields that the low-frequency region of $\delta\Delta_1$ is at frequencies such that an additional requirement is satisfied:

$$\nu\gamma(\omega/\omega_0) < \tfrac{1}{2}(1-\gamma/2\omega_0\tau_E)^2 . \qquad (21)$$

When these conditions hold, one obtains

$$(k_1\delta_L)^2 = 2\kappa^2 , \qquad (22)$$

$$(k_2\delta_L)^2 = 2\kappa^2\left(\frac{\gamma}{2\omega_0\tau_E} - i\gamma\frac{\omega}{\omega_0} - i\gamma\frac{\omega}{\omega_0}\frac{\nu}{2}\frac{1}{1-\gamma/2\omega_0\tau_E}\right), \qquad (23)$$

$$k_1 a_1 = -2ka\left\{1 + i\gamma\frac{\omega}{\omega_0}\frac{\nu}{2}\left[\left(1-\frac{\gamma}{2\omega_0\tau_E}\right)\left(i\gamma\frac{\omega}{\omega_0} + \frac{(2k\delta_L)^2}{2\kappa^2} - \frac{\gamma}{2\omega_0\tau_E}\right)\right]^{-1}\right\}, \qquad (24a)$$

$$k_2 a_2 = 2ka\, i\gamma\frac{\omega}{\omega_0}\frac{\nu}{2}\left[\left(1-\frac{\gamma}{2\omega_0\tau_E}\right)\left(i\gamma\frac{\omega}{\omega_0} + \frac{(2k\delta_L)^2}{2\kappa^2} - \frac{\gamma}{2\omega_0\tau_E}\right)\right]^{-1} . \qquad (24b)$$

Using condition (20), which insures that $(k\delta_L)^2 \sim 1$, together with the requirement (21), one can check that as long as

$$\left|\left(1-\frac{\gamma}{2\omega_0\tau_E}\right)\Big/\left(i\gamma\frac{\omega}{\omega_0} - \frac{\gamma}{2\omega_0\tau_E} + \frac{(2k\delta_L)^2}{2\kappa^2}\right)\right| \lesssim 1 , \qquad (25)$$

$$\kappa^2 \gtrsim 1, \qquad (26)$$

then the a_1, a_2 terms in expression (12) for $\delta\Delta_1$ are negligible compared to the a term. Namely, conditions (21), (25), and (26) determine the frequency region in which the order parameter follows spatially the field. In this region, the third-harmonic component takes the form

$$H_1(Z=0) = \frac{H_0^3}{\Delta\delta_L^2}\,\frac{a/A_0^2}{k^4(\omega)[3+k(3\omega)/k(\omega)]} , \qquad (27)$$

where a is given in (15) and H_0 denotes the amplitude of the applied magnetic field.

To proceed, one needs to know the values of the parameters ν and γ appearing in (27), which depend on the pair-breaking parameter ρ [see Eqs. (14)]. In the Appendix we calculate ν in three limiting cases:

(i) gap regime, $\Delta \gg \rho 2\pi T$,

$$\nu = [2\pi^3/7\zeta(3)](T/\Delta) , \qquad (28a)$$

in which $\zeta(3)$ is the Rieman ζ function;

(ii) gapless regime, $\Delta \ll \rho 2\pi T$, $\rho \ll 1$, which occurs in dilute alloys close enough to T_c,

$$\nu = [\pi^2/14\zeta(3)](1/\rho) ; \qquad (28b)$$

(iii) gapless superconductors, $\rho \gg 1$,

$$\nu = 4/\rho^2 . \qquad (28c)$$

In the first two cases $\nu \gg 1$ while for gapless superconductors, ν is negligibly small. On the other hand, γ [see Eqs. (4) and (14)] is of the order 1 both for $\rho \ll 1$ and $\rho \gg 1$:

$$\gamma \simeq \begin{cases} [\pi^4/14\zeta(3)], & \rho \ll 1 \\ 12 , & \rho \gg 1. \end{cases} \qquad (29)$$

The fact that ν is large in dilute alloys and very small in gapless superconductors leads to a crucial difference between the two types of materials. In gapless superconductors, condition (21), which marks the low-frequency behavior of the order parameter, is automatically satisfied. As a result, there is only one characteristic frequency in the system, namely, ω_0. In this case we recover for $H_1(Z=0)$ the Gorkov-Eliashberg[2] result

$$H_1(Z=0) = \frac{1}{4}\frac{H_0^3}{H_c^2}\left[[k(\omega)\delta_L]^4\left(3+\frac{k(3\omega)}{k(\omega)}\right)\times\left(i\gamma\frac{\omega}{\omega_0} + \frac{(2k\delta_L)^2}{2\kappa^2} - 1\right)\right]^{-1} . \qquad (30)$$

(Note that in Ref. 2, $(2k\delta_L)^2/2\kappa^2$ was neglected compared to 1.) This yields that the relaxation time of the order parameter in gapless superconductors is $\sim(\Delta^2\tau_S)^{-1}$. On the other hand, in the cases where $\nu \gg 1$, the low-frequency behavior of $\delta\Delta_1$ is marked by a frequency ω_1 which is much smaller than ω_0. From (21) and (24), we have

$$\omega_1 \simeq \omega_0/\gamma\nu \simeq \begin{cases} (\Delta/T)\omega_0, & \text{gap regime} \\ \rho\omega_0 , & \text{gapless regime } (\rho \ll 1). \end{cases} \qquad (31)$$

[Here we have assumed $\gamma/2\omega_0\tau_E \lesssim 1$, a condition probably satisfied in the experiment[1] (see discussion below).] Note the difference in the temperature dependence in the two cases: In the gap regime $\omega_1 \sim (T_c - T)^{3/2}$; in the gapless regime (closer to T_c) $\omega_1 \sim T_c - T$. For $\nu \gg 1$ the third-harmonic amplitude becomes

$$H_1(Z=0)=\frac{1}{4}\frac{H_0^3}{H_c^2}\left\{[k(\omega)\delta_L]^4\left(3+\frac{k(3\omega)}{k(\omega)}\right)\left(i\gamma\frac{\omega}{\omega_0}+\frac{(2k\delta_L)^2}{2\kappa^2}-1-\frac{i\gamma(\omega/\omega_0)(\nu/2)}{i\gamma(\omega/\omega_0)+[(2k\delta_L)^2/2\kappa^2]-(\gamma/2\omega_0\tau_E)}\right)\right\}^{-1}. \qquad (32a)$$

The third-harmonic output power is given by[1]

$$P \propto \omega[|H_1(Z=0)|/|k(3\omega)|]^2, \qquad (32b)$$

where the constant of proportionality is determined by the experimental apparatus parameters. Equation (32b) is depicted in Fig. 1 for $\rho=0$, 1, and 10. To draw the figure, we have denoted $\omega/\omega_0 = c_0/(1-T/T_c)$, where

$$c_0=\frac{\omega}{4\pi T}\psi'(\tfrac{1}{2}+\rho)/\gamma[1-\rho\psi'(\tfrac{1}{2}+\rho)].$$

This yields $\omega/4\pi T_c = 0.03$, 0.03, and 0.003 for $\rho=0,1$, and 10, respectively. As the ratio $\omega/4\pi T_c$ is decreased, the peak moves towards $T/T_c=1$ and becomes narrower. In three cases depicted in Fig. 1 $c_0=0.03$, 0.04, and 0.03 for $\rho=0,1$, and 10, respectively.

To study the consequences of expression (32), we approximate in it $k(\omega)\delta_L \sim 1$. Then, using (21) and (25) it becomes

$$H_1(Z=0)=\frac{H_0^3}{16H_c^2}\left\{-1+\frac{2}{\kappa^2}+\frac{(\gamma\omega/\omega_0)^2}{-1+(2/\kappa^2)}\left[1-\tfrac{1}{2}\nu\frac{(2/\kappa^2)-(\gamma/2\omega_0\tau_E)}{(2/\kappa^2)-(\gamma/2\omega_0\tau_E)^2+(\gamma\omega/\omega_0)^2}\right]^2\right\}^{-1}. \qquad (33)$$

We can distinguish here between two cases. (i) $2/\kappa^2 > \gamma/2\omega_0\tau_E$, $\kappa^2\nu > 4$. In this case the phonon inelastic scattering time is omitted and as a result the relaxation time of the order parameter is

$$\frac{\gamma\nu\kappa^2}{4\omega_0} \simeq \begin{cases}\kappa^2 T/\Delta\omega_0, & \text{gap regime}\\ \kappa^2/\omega_0, & \text{gapless regime}.\end{cases} \qquad (34)$$

This result, for the gapless regime, was used in Ref. 1 for analyzing the experimental data. The temperature dependence of the relaxation time is again different in the two regions. (ii) $2/\kappa^2 < \gamma/2\omega_0\tau_E$. In this case τ_E plays an important role, and the relaxation time is $\nu\tau_E$. This does not depend on the temperature in the gapless regime, but in the gap regime it becomes $2\pi^3 T\tau_E/7\zeta(3)\Delta$, with temperature dependence $(T_c-T)^{-1/2}$. It is interesting to note that it is the same result obtained in Ref. 5 for the decay time of the order parameter in the longitudinal mode. The longitudinal mode is associated with fluctuations in the order parameter amplitude, i.e., the fluctuations caused by the field in the present case. Note that this result is reached close to T_c, where ω_0 becomes small (but there is still a gap in the excitation spectrum). As ω_0 is the upper limit on the frequency, this region might be difficult to attain experimentally.

IV. DISCUSSION

An expression for the third-harmonic amplitude, generated in a superconductor by a weak magnetic field, was derived. We have operated in the dirty limit and included in the calculations pair-breaking effect and inelastic scattering of the electrons by the phonons.

It was found that depending on the strength of the depairing parameter, there are two characteristic frequencies in this problem. This first, ω_0, marks the low-frequency behavior of the field, i.e., the region in which the field penetrattion is determined by the Meissner effect. This frequency is $\sim\Delta^2/T$ for small values of ρ, and $\Delta^2\tau_S$ in gapless

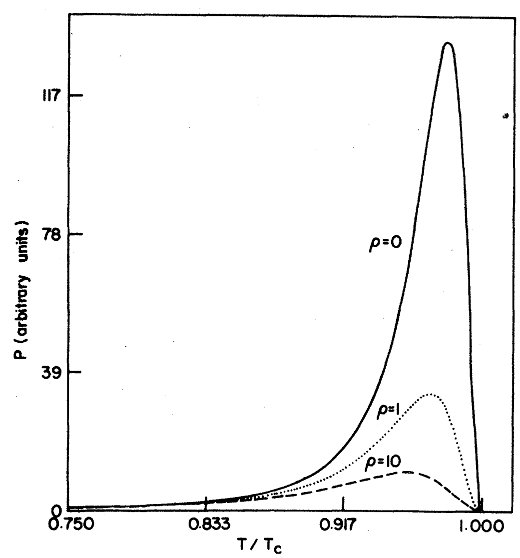

FIG. 1. Third-harmonic output power in arbitrary units. Here $k^2 = 50$, $\omega\tau_E = 10$ and $\omega/4\pi T_c = 0.03$, 0.03, and 0.003 for $\rho = 0$, 1, and 10, respectively.

superconductors ($\rho \gg 1$). The second, ω_1 determines the low-frequency behavior of the deviation of the order parameter from equilibrium. In gapless superconductors, it turns out that $\omega_0 \sim \omega_1$. For $\rho \ll 1$, $\omega_1 < \omega_0$. Summarizing, we have

$$\omega_1 \sim \frac{\Delta^2}{T}\left(\frac{\Delta}{T}\right), \quad \text{gap regime},$$

$$\omega_1 \sim \frac{\Delta^2}{T}(\rho), \quad \text{gapless regime } (\rho \ll 1), \tag{35}$$

$$\omega_1 \sim \frac{\Delta^2}{T}(\rho^{-1}), \quad \text{gapless superconductors } (\rho \gg 1).$$

In all three cases Δ^2/T is reduced by a small factor. These frequencies are quite important, since the amplitude of the measured effect (third-harmonic power output) is proportional to the frequency.

The inclusion of the inelastic phonon scattering time τ_E leads to an interesting result. We have assumed throughout the calculation that $\omega_0\tau_E \gg 1$. This is quite plausible, since the frequency used in the experiment[1] is ~10^{10} Hz. But we have found that for $\omega_0\tau_E < \kappa^2$, which may occur for large values of the Landau-Ginzburg parameter, and close enough to T_c, the relaxation time of the order parameter is ~$\tau_E T/\Delta$. This may be used to extract explicit value for τ_E.

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APPENDIX: CALCULATION OF I

From the theory for nonstationary superconductors (see Refs. 2–6), it is found that

$$I = \frac{1}{2\Delta T}\int \frac{d\epsilon}{4\pi i}\,\frac{1}{\cosh^2(\epsilon/2T)}\,\frac{\beta_2^2(\epsilon)}{\beta_1(\epsilon)}, \tag{A1}$$

in which

$$\beta_2 = \frac{\pi i}{2}\left(\frac{\Delta_R}{W_R} + \frac{\Delta_A}{W_A}\right), \quad \beta_1 = \frac{\pi i}{2}\left(\frac{\epsilon_R}{W_R} - \frac{\epsilon_A}{W_A}\right), \tag{A2}$$

$$W_{R,A}^2 = \epsilon_{R,A}^2 - \Delta_{R,A}^2,$$

and $\epsilon_{R,A}\Delta_{R,A}$ are the retarded (advanced) energy and order parameter "dressed" by the various scattering mechanisms.[2-4]

In the gap regime, $\Delta \gg \rho 2\pi T$,

$$\beta_2 \simeq \pi i\,\frac{\Delta}{(\epsilon^2 - \Delta^2)^{1/2}}, \quad \beta_1 \simeq \pi i\,\frac{|\epsilon|}{(\epsilon^2 - \Delta^2)^{1/2}}. \tag{A3}$$

The important contribution to the integral in (A1) comes from $\epsilon \simeq \Delta$, and thus we may approximate $\cosh^2\epsilon/2T$ by 1 and obtain

$$I = \pi/8T. \tag{A4}$$

When $\Delta \ll \rho 2\pi T$,

$$\beta_2 \simeq \pi i\,\frac{\Delta\epsilon}{\epsilon^2 + (2\pi T\rho)^2}, \quad \beta_1 \simeq \pi i, \tag{A5}$$

and as a result

$$I = \frac{\Delta}{(4\pi T)^2}\left[\frac{1}{\rho}\psi'(\tfrac{1}{2}+\rho) + \psi''(\tfrac{1}{2}+\rho)\right]. \tag{A6}$$

In the gapless regime of dilute alloys ($\rho \ll 1$), this yields

$$I \simeq [\Delta/(4\pi T)^2][\psi'(\tfrac{1}{2}+\rho)/\rho] \tag{A7}$$

and for gapless superconductors, $\rho \gg 1$,

$$I \simeq [\Delta/(4\pi T)^2](2/3\rho^4). \tag{A8}$$

Inserting the values of I into Eq. (14c) for ν and using Eq. (4b), we find

$$\nu = \frac{\pi}{8T}\,\frac{2}{\Delta}\,\frac{1}{\beta_0} = \frac{2\pi^3}{7\zeta(3)}\,\frac{T}{\Delta}, \quad \text{gap regime}, \tag{A9}$$

in which ζ is Rieman zeta function;

$$\nu = \frac{\Delta}{(4\pi T)^2}\,\frac{\psi'(\frac{1}{2}+\rho)}{\rho}\,\frac{2}{\Delta\beta_0} \simeq \frac{\pi^2}{14\zeta(3)}\,\frac{1}{\rho}, \quad \text{gapless regime}, \rho \ll 1, \tag{A10}$$

$$\nu = \frac{\Delta}{(4\pi T)^2}\,\frac{2}{3\rho^4}\,\frac{2}{\Delta\beta_0} \simeq \frac{4}{\rho^2}, \quad \text{gapless superconductors}, \rho \gg 1. \tag{A11}$$

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