Phonon and Quasiparticle Dynamics in Superconducting Aluminum Tunnel Junctions*

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(Received 28 July 1975)

The effect of the 2Δ recombination phonons in modifying the effective lifetime τ_{eff} for quasiparticle recombination has been determined for the first time in a double-junction experiment by comparing measurements taken both with and without a helium film covering the junctions. For a double junction made from evaporated aluminum films 300 Å thick deposited on a glass substrate, we find $\tau_{eff}/\tau_R = 8.6 \pm 1.1$ in the absence of the helium film, and, at $\Delta/kT = 6$, $\tau_R = 1.4 \pm 0.1$ μsec.

In a superconductor, quasiparticles decay to the ground state by forming pairs, and the energy released in this process, called recombination, is carried away by phonons[1] of energy $\hbar\omega \geq 2\Delta$. These recombination phonons can modify the experimentally observed lifetimes for this process by breaking pairs and thus creating new quasiparticles, rather than simply escaping from the superconductor.[2] Several investigators have tried to account for this effect in superconducting films by comparing samples which differed in some parameter affecting the phonon escape rate, such as the film thickness[3,4] or the substrate onto which the films were evaporated,[3,5] but their results have been inconclusive, except to verify the importance of this effect. This experiment uses the double-junction method[5] for measuring lifetimes, with the additional feature that the phonon escape rate can be changed *in situ* by covering the junctions with a helium film. Furthermore, a second source of 2Δ phonons, generated by the relaxation of excited quasiparticles of energy $E \geq 3\Delta$, obviates the need for any theoretical description of the nature of the phonon escape. With this method, the magnitude of the effect of the recombination phonons, and hence the intrinsic quasiparticle lifetime, can be directly determined.

The double junction consists of three superimposed superconducting films separated by two tunneling barriers, forming two tunnel junctions. One junction, called the generator, is biased at a voltage $|eV| \geq 2\Delta$, where it creates quasiparticles in its two films. The other junction, called the detector, is biased at a voltage less than 2Δ, where the tunneling current measures the number of quasiparticles in its two films. Using the method of Rothwarf and Taylor,[2] we have resolved the steady-state equations describing the phonons and the quasiparticles in the double junction by analyzing each of the three films separately. This is necessary because, while quasiparticles are *directly* injected only into the two generator films, they are produced *indirectly* in all three films through the action of the recombination phonons. Details of the calculation will be presented in a future paper. In the "linear regime," we obtain the same result for the effective lifetime $\tau_{eff,1}$:

$$\tau_{eff,1} \equiv \frac{\partial(n_1+n_2)}{\partial I_0} = \tau_R(1+\tau_\gamma/\tau_1), \quad (1)$$

where n_1, n_2 are the excess numbers of quasiparticles per unit volume in the two detector films, I_0 is the rate at which quasiparticles are created in each of the two generator films, τ_R is the recombination lifetime, and τ_1 is the average lifetime for a phonon of energy $\hbar\omega \geq 2\Delta$ to break a pair.[6] τ_γ is a lifetime parametrizing phonon escape, and depends on the acoustic coupling of the films to the substrate, to a helium film (if present), and to each other. In the nonlinear regime, where τ_{eff} is a function of n_1 and n_2, our result is equivalent to that of Rothwarf and Taylor only if $n_1 = n_2$ or $n_1 = 0$. This is true for τ_γ much greater or much less than τ_1, and corresponds to most or none of the excess quasiparticle population being produced from the recombination phonons. In this limit, and for small deviations from linearity, we find

$$\tau_{eff,nl} \approx \tau_{eff,l}[1-(n_1+n_2)/2N_T], \quad (2)$$

where N_T is the thermal number of quasiparticles in the junctions per unit volume.

In terms of experimentally measured quantities, we have[7]

$$\tau_{eff,1} = e\Omega f\left(\frac{N_T}{I_{det,T}}\right)\frac{\partial I_{det}}{\partial I_{gen}}\left(1+f\frac{\Delta I_{det}}{I_{det,T}}\right), \quad (3)$$

where Ω is the volume of the two generator films, $\Delta I_{det} = I_{det} - I_{det,T}$ is the change in the detector current from its thermal value, I_{gen} is the generator current, and f is a geometrical factor describing the overlap of the junctions.

When the generator junction is biased at a voltage $|eV| \geq 4\Delta$, some of the created quasiparticles will have an energy $E \geq 3\Delta$ and may relax to the gap by emitting a phonon of energy $\hbar\omega \geq 2\Delta$. This source of phonons has the form

$$\delta\dot{n}_\omega = K(V)[I_0 - I_0(4\Delta)], \quad |eV| \geq 4\Delta, \tag{4}$$

where $\delta\dot{n}_\omega$ is the rate at which 2Δ phonons are generated in each of the two generator films per unit volume, $I_0(4\Delta)$ is the value of I_0 at $|eV| = 4\Delta$, and $K(V)$ is a constant ($=0$ for $|eV| < 4\Delta$) which may depend on voltage, and in principle can be calculated theoretically. This effect has been studied by several investigators in experiments using separate tunnel junctions as phonon generators and detectors.[8-14] Some find that $K(V)$ is sharply peaked at 4Δ,[10-12,14] while others[9,13] find that it depends only weakly on voltage above 4Δ.

The effective lifetime at $|eV| \geq 4\Delta$ becomes[15]

$$\tau_{\mathrm{eff},1} = \tau_R\{1 + (\tau_\gamma/\tau_1)[1 + 2K(V)]\} \tag{5}$$

and the nonlinear correction (2) still applies. The jump in τ_{eff} which occurs at 4Δ, which we call $G(V)$, is given by

$$\begin{aligned} G(V) &\equiv \tau_{\mathrm{eff},1}(|eV| > 4\Delta)/\tau_{\mathrm{eff},1}(|eV| < 4\Delta) \\ &= 1 + 2K(V)[\tau_\gamma/(\tau_1 + \tau_\gamma)]. \end{aligned} \tag{6}$$

Call τ_{wet} (τ_{dry}) the value of $\tau_{\mathrm{eff},1}$ at a generator voltage $|eV| < 4\Delta$ in the presence (absence) of helium. Measurement of τ_{wet}, τ_{dry}, $G_{\mathrm{wet}}(V)$, and $G_{\mathrm{dry}}(V)$ allows us to determine $\tau_{\mathrm{dry}}/\tau_R$, and hence τ_R, from Eqs. (1) and (6):

$$\frac{\tau_{\mathrm{dry}}}{\tau_R} = 1 + \left[\frac{G_{\mathrm{dry}}(V) - 1}{G_{\mathrm{dry}}(V) - G_{\mathrm{wet}}(V)}\right]\left[\frac{\tau_{\mathrm{dry}} - \tau_{\mathrm{wet}}}{\tau_{\mathrm{wet}}}\right]. \tag{7}$$

Note that this result is independent of $K(V)$.

We shall now discuss the assumptions involved in the derivation of Eq. (7). We have assumed, first, that no physical process, other than the relaxation of injected quasiparticles by emission of phonons of energy $\hbar\omega \geq 2\Delta$, can explain, in part or in total, the jump in τ_{eff} which occurs at $|eV| = 4\Delta$; and second, that the constant $K(V)$ is unaffected by the presence or absence of the helium film. We shall also explain how the quantity $G(V)$ is obtained from the actual experimental data.

With regard to the first assumption, Lancashire[16] has proposed that quasiparticles of energy $E \geq 3\Delta$ may relax to the gap by pair breaking at a rate, in aluminum, comparable to decay by phonon emission. This would add a term $K'(V) \times [I_0 - I_0(4\Delta)]$ to the steady-state equations for each of the two generator films, and the new result for $G(V)$ would be enhanced by exactly $K'(V)$. Since this process results in three quasiparticles near the gap, $K'(V)$ should be very sharply peaked at $|eV| = 4\Delta$.[17] None of the published data for aluminum[7,13] shows this peak where it cannot be attributed to nonlinear effects; hence we may safely conclude that the predominant relaxation process in aluminum is by phonon emission, or, alternatively, that $\alpha(V) \equiv K'(V)/[K'(V) + 2K(V)]$ is small. Data for tin[11,14] and lead[10] which do exhibit a peak at $|eV| = 4\Delta$, could be interpreted as evidence for this type of decay in those metals.

$\alpha(V)$ enters into our derivation of $\tau_{\mathrm{dry}}/\tau_R$ as

$$\tau_{\mathrm{dry}}/\tau_R = (\tau_{\mathrm{dry}}/\tau_R)_{\mathrm{exp}}[1 - \alpha(V)],$$

where $(\tau_{\mathrm{dry}}/\tau_R)_{\mathrm{exp}}$ is the value obtained from the data if $\alpha(V)$ is assumed to be zero. Therefore, as long as $\alpha(V)$ is small, we can safely neglect this effect.

With regard to the second assumption, Kinder and Dietsche[18] have found that phonons scattered off a helium film may be shifted either up or down in frequency. This would have two effects on the double-junction experiment when the helium film is present. First, it would reduce τ_γ for the phonons produced in the recombination process, since some would be scattered out of range $\hbar\omega \geq 2\Delta$. This would not affect our analysis since τ_γ is just a parameter about which we make no assumptions and which does not enter explicitly into our final result. Second, $K(V)$ would be effectively smeared out, since it is a measure of the rate at which relaxation phonons with energies $\hbar\omega \geq 2\Delta$ are produced. Fortunately, the importance of this effect can be tested by comparing the shapes of τ_{dry} and τ_{wet} versus I_{gen} both above and below $|eV| = 4\Delta$: If the curves are not similar in either region, the effect is important. In this case, and because there may be other structure in τ_{eff} due to effects associated with the energy distribution of the quasiparticles or phonons,[11] or to gap anisotropy,[12] G must be labeled as $G(V_>, V_<)$, where $V_>$ and $V_<$ are the generator voltages above and below $4\Delta/e$ used in the measurement. Insofar as it may affect the determination of $\tau_{\mathrm{eff}}/\tau_R$, the only restriction of $V_>$ and $V_<$ is that they be chosen in regions where the shapes of the curves do not change in the presence or absence of helium.

Our double junctions were of the type Al-oxide-Al-oxide-Al, where the aluminum films were evaporated onto a fire-polished glass slide at rates ≈ 5 Å/sec at pressures $< 10^{-5}$ Torr, and were of equal thickness. The oxide layers were grown by exposing the films to wet oxygen for various lengths of time. The resistances of the detector junctions are typically ≈ 1 Ω, and the generator junctions ≈ 1000 Ω, where the junction area is ≈ $\frac{1}{2}$ mm^2. All the experiments were performed in a ^{3}He refrigerator in magnetic fields < 0.01 G normal to the films and ≈ 20 G parallel to the films. Helium could be admitted to or removed from the sample chamber through a capillary tube, and the film thicknesses, as determined from measurements of third-sound velocity, were greater than eight atomic layers, which was thick enough that the addition of more helium had no noticeable effect on τ_{eff}.

Figure 1 is an experimental graph of $\partial I_{det}/\partial I_{gen}$, taken at a temperature $T = 0.41$ K, for a sample made from 400-Å-thick films. Two curves are displayed: The full curve, taken in the presence of a helium film, has a vertical sensitivity twice that of the dashed curve, taken in the absence of helium. Both curves show the change of slope at 6Δ and the peak at ⩾2Δ which are of the same origin as the structures seen in the experiments using separate tunnel junctions as phonon detectors and generators. The full curve has a sharper tail at voltages just below 4Δ and a more pronounced dip at a voltage ≈ 2.5Δ. Both of these structures are probably due to the phonon frequency-conversion process mentioned above. Most importantly, the jump at 4Δ is noticeably different in the two curves.

$G_{wet}(V_>, V_<)$ and $G_{dry}(V_>, V_<)$ are calculated at voltages $V_> = 5\Delta/e$ and $V_< = 3\Delta/e$, where the shapes of the two curves are similar and the effects of the finite modulation current δI_{gen} are unimportant. τ_{dry} and τ_{wet} are measured at a generator current about halfway up the jump at 2Δ. All measurements are taken at both positive and negative generator currents and detector biases of ± 100 μV, and then averaged. The measurements were found to be reproducible from day to day.

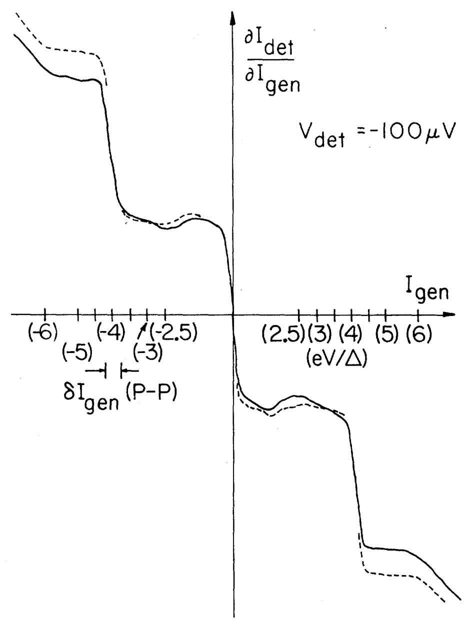

FIG. 1. $\partial I_{det}/\partial I_{gen}$ versus I_{gen} for sample No. 2 at $T = 0.41$ K. The corresponding generator voltages eV/Δ are in parentheses. Two curves are shown: The full curve was taken with a helium film covering the junctions, and has twice the sensitivity of the dashed curve, taken in the absence of helium. See text.

Our data for three samples are summarized in Table I. τ_{dry} and τ_{wet} both show the expected temperature dependence, $\sim N_T^{-1}$, and are similar in magnitude to other published results.[3,7] Systematic errors in the lifetimes due to, for example, uncertainties in the junction geometry could be as high as 25%; these would not, however, affect the observed temperature depen-

TABLE I. Summary of experimental results.

Sample	Film thickness (Å)	2Δ (μV)	T (K)	τ_{dry} (μsec)	τ_{wet} (μsec)	G_{dry}	G_{wet}	τ_{dry}/τ_R
1	300	390	0.42	6.59	4.08	2.93	2.73	7.0
			0.40	9.17	5.46	2.98	2.82	9.4
			0.38	12.43	7.69	3.01	2.86	9.3
						Average:		8.6 ± 1.1
2	400	394	0.45	3.91	1.82	2.88	2.63	9.6
			0.41	6.10	2.95	2.92	2.64	8.3
3	400	393	0.45	4.47	2.01	2.86	2.54	8.1

dence or the ratio $\tau_{\rm dry}/\tau_R$. The estimated uncertainties for $\tau_{\rm dry}/\tau_R$ are on the order of 10–15%; this agrees well with the statistical error calculated for the 300-Å sample based on the assumption that $\tau_{\rm dry}/\tau_R$ is independent of temperature. Its value for this sample, 8.6 ± 1.1, is consistent with recent estimates by Long,[19] but much greater than earlier calculations.[5,7] Also, the magnitude of the deduced intrinsic lifetimes, $\tau_R = 1.4 \pm 0.1$ μsec at $\Delta/kT = 6$, is significantly smaller than the theoretical prediction of Gray.[20]

A more complete description and other aspects of this experiment will be published elsewhere.

*Research supported in part by the National Science Foundation under Grants No. DMR 72-03026 and No. GH-37892.

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[6]Our definition of $\tau_{\rm eff}$ actually differs from that of Rothwarf and Taylor who use the ratio instead of the derivative. Also, our $\tau_1 = 2/\beta$, where β is the transition probability for the breaking of pairs by phonons.

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COMMENTS

Microscopic Theory and Deformation-Dipole Model of Lattice Dynamics

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(Received 25 August 1975)

It is shown that a deformation-dipole model more general than that of Hardy follows rigorously from Zeyher's microscopic theory of lattice dynamics of ionic crystals. In addition to the self-energy correction term due to the electric dipoles in Hardy's model, Zeyher's theory gives a similar term due to the deformation dipoles. The general shell model corresponds to a special form of the new deformation-dipole model.

The two commonly used and quite realistic phenomenological models of lattice dynamics of ionic crystals are the deformation-dipole model (DDM) and the shell model.[1] Almost all the microscopic calculations so far have been compared with the shell model only. Recently, Zeyher has developed a microscopic theory of lattice dynamics of ionic crystals in terms of highly localized nonorthogonal