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Quasiparticle Lifetimes and Microwave Response in Nonequilibrium Superconductors*

A. Rothwarf,† G. A. Sai-Halasz, and D. N. Langenberg
Department of Physics and Laboratory for Research on the Structure of Matter, University of Pennsylvania, Philadelphia, Pennsylvania 19174
(Received 21 March 1974)

A theoretical treatment is presented of the quasiparticle-recombination dynamics of a nonequilibrium superconductor. The microwave reflectivity of a superconducting film in the presence of external pair breaking is calculated. It is shown how microwave experiments may permit the separation of the effective recombination time into its intrinsic and recombination-phonon components.

Testardi[1] has shown experimentally that when a thin superconducting film is driven normal by intense pulsed optical radiation, the transition displays characteristics which cannot be accounted for by simple lattice heating. He suggested that excess quasiparticles created by photon-induced pair breaking were responsible. Motivated by this observation, Owen and Scalapino[2] investigated a modified BCS model of a superconductor in which the quasiparticle density is maintained at a level above the thermal-equilibrium value by an external source of dynamic pair breaking. Among the predictions of this model are a dependence of the energy gap on the excess quasiparticle density, and a first-order transition to the normal state at a critical excess quasiparticle density. Some of the model's predictions have been experimentally confirmed by Parker and Williams[3] using tunnel junctions irradiated with laser light.

Some time ago, Rothwarf and Taylor[4] pointed out the crucial importance of recombination phonons in the coupled quasiparticle-pair–phonon system in a nonequilibrium superconductor. The interpretation of nonequilibrium experiments using tunnel junctions depends on assumptions about the behavior of these phonons in the relatively complex junction structure. It would obviously be desirable to test the Owen-Scalapino model in a simple thin film, in which the phonons can be accounted for with greater certainty. The use of microwave-frequency reflection, absorption, or transmission measurements as a probe of the quasiparticle density makes this possible. The quasiparticle density can be monitored in the superconducting state in contrast to the quasi-dc experiment of Testardi, in which only the transition to the normal state could be studied. Furthermore, experiments of this type offer the possibility of direct time-resolved studies of quasiparticle dynamics if the relevant times are sufficiently long. Such experiments are reported in the following Letter.[5] In this Letter we present the theoretical framework required for interpretation of the experiments.

We first require a connection between the quasiparticle density and an observable microwave parameter. In the Owen-Scalapino model, it is assumed that the quasiparticles and phonons are in thermal quasiequilibrium at temperature T, but that the quasiparticles are not in chemical equilibrium with the pairs. The distribution function of the quasiparticles is

$$f(E,T,n)=\{\exp((kT)^{-1}[E-\mu^*(T,n)])+1\}^{-1}, \quad (1)$$

where E is the quasiparticle energy, μ^* is the quasiparticle chemical potential, and n is the quasiparticle density excess over the thermal-equilibrium value at temperature T, measured in units of $4N(0)\Delta_0$. $N(0)$ is the single-spin density of states at the Fermi level, and Δ_0 is the BCS gap parameter at zero temperature and in equilibrium ($n=0$), with the pair chemical potential $\mu=0$. Both E and μ^* depend on the gap parameter, which in turn depends on T and n. We have used the formulas given by Mattis and Bardeen,[6] modified by inserting the distribution function Eq. (1) and the Owen-Scalapino results for $\Delta(T,n)$ and $\mu^*(T,n)$, to calculate the real and imaginary parts of the normalized conductivity, $\sigma_1(T,n)/\sigma_N$ and $\sigma_2(T,n)/\sigma_N$. Using these, we have calculated the normal-incidence microwave reflectivity of a film-substrate combination. An example of the results is shown in Fig. 1.

Next we need a connection between n and the intensity of the external pair-breaking source, e.g., incident light intensity. This link is provided by recombination dynamics. Rothwarf and Taylor[4] in their work assumed that the phonon pair-breaking probability $\beta/2$ and the quasiparticle recombination coefficient R are independent of the number of excess quasiparticles. However, these parameters depend on the gap parameter, and hence, in general, on n. We assume that each depends on n only through its explicit dependence on Δ. With this assumption and from the rate equations[4,7] for the equilibrium case, we find the ratio $\beta/R=N_T^2(T,\Delta(T,n))/N_{\omega T}(T,\Delta(T,n))$, where N_T is the thermal-equilibrium quasiparticle density and $N_{\omega T}$ is the thermal-equilibrium density of phonons with energy greater than 2Δ, each for

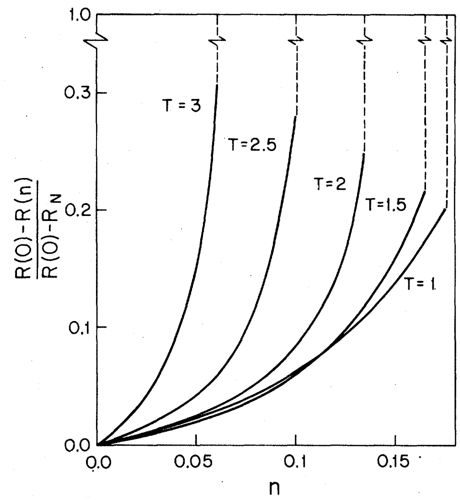

FIG. 1. Normalized change in microwave reflectivity (70 GHz) of a 500-Å-thick Sn film as a function of normalized excess quasiparticle density, with temperature as a parameter in Kelvin degrees. R_N is the normal-state reflectivity, $R(0)$ is the superconducting-state reflectivity, and $R(n)$ is the reflectivity of the state with n excess quasiparticle density. The vertical line at the right-hand end of each curve marks the first-order transition to the normal state predicted by the Owen-Scalapino model (Ref. 2).

given T and $\Delta(T,n)$. If, as assumed by Owen and Scalapino, the quasiparticle and phonon distributions can be characterized by the same temperature T, then for $kT/\Delta \ll 1$, β/R is simply proportional to $\Delta^{-1}(T,n)$, and is otherwise temperature independent.

With this change, from the rate equations[4,7] for the steady state or quasiequilibrium, we get

$$N^2=N_T^2(T,n)+I_0\left(\frac{1}{R}+\frac{N_T^2(T,n)}{2N_{\omega T}(T,n)}\tau_\gamma\right), \quad (2)$$

where $N=N_T(T,n)+\Delta N$ is the total quasiparticle density, $\Delta N=n[4N(0)\Delta_0]$, I_0 is the quasiparticle injection rate, and τ_γ^{-1} is the net transition probability for loss of phonons from the energy range $>2\Delta$ through processes other than pair breaking.

We now suppose that the external pair-breaking source is light and that the film absorbs energy flux P. We assume that in quasiequilibrium a fraction F of this energy is converted to quasiparticles. Then $I_0=FP/d\bar{E}_{qp}$, where d is the film thickness and $\bar{E}_{qp}$ is the average quasiparticle energy. The factor F can be estimated with the assumption that the high-energy quasiparticles created by an incident photon decay by the emission of phonons. The phonons emitted will be predominantly those corresponding to peaks in the phonon density of states. These phonons in turn can create quasiparticles by breaking pairs. These quasiparticles will tend to fall to the gap edge by the emission of phonons because of the large density of states at the gap. If the emitted phonon can further break pairs, the process continues until the emitted phonons have an energy less than 2Δ. The conversion efficiency can thus be obtained from the ratio of the energies at peaks in the phonon density of states to the energy gap. In Pb we find conversion efficiencies of ~0.6, 0.7, and 0.8 for the two transverse phonon peaks and the longitudinal peak, respectively. Since we expect the transverse peaks to be more effective as a result of the predominance of the umklapp scattering in Pb, a value of $F \approx \frac{2}{3}$ is reasonable for Pb. For Sn, taking into consideration all peaks in the phonon spectrum, we get $F \approx \frac{3}{4}$.

Other loss mechanisms such as the emission of phonons with energy less than 2Δ, during the high-energy quasiparticle bremsstrahlung, or the creation of low-energy phonons from high-energy ones by inelastic processes, account for a negligible portion of the energy.

The rate at which the high-energy phonons will create quasiparticles can be estimated from the fact that the 2Δ phonon lifetime against pair breaking is at least a factor 10 smaller than the quasiparticle lifetime, even when the latter is sharply reduced at large excess quasiparticle densities. Consequently almost all the energy resides in the quasiparticle component of the system. This conclusion is also supported by direct calculations of the total excess quasiparticle energy, $\Delta N\bar{E}_{qp}$, and the total excess phonon energy, $\Delta N_\omega \bar{E}_\omega$, based on Rothwarf and Taylor's equations. For any reasonable set of parameter values, $\bar{E}_{qp}$ is very nearly Δ, $\bar{E}_\omega$ is very nearly 2Δ, and $\Delta N_\omega \ll \Delta N$. F depends on n through $\Delta(n)$ in general, but is nearly always close to unity. Hence we can write

$$I_0 = FP/d\Delta(T,n). \tag{3}$$

Equations (2) and (3) together give the required connection between absorbed light power P and the excess quasiparticle density n. The microwave reflectivity as a function of P will therefore give information about recombination dynamics.

An effective quasiparticle lifetime τ_{eff} can be defined in quasiequilibrium by $\Delta N = I_0 \tau_{\text{eff}}$.[7] From Eq. (2),

$$\tau_{\text{eff}} = \left(\frac{1}{R} + \frac{N_T^2(T,n)}{2N_{\omega T}(T,n)}\tau_\gamma\right)(\Delta N + 2N_T)^{-1}. \tag{4}$$

The coefficient of τ_γ varies as $1/\Delta(n)$ for $kT \ll \Delta$, making the role of the recombination phonons even more important than in Rothwarf and Taylor's original work. In the strong-injection regime, $\Delta N \gg N_T$, and τ_{eff} can become very short, making time-resolved experiments very difficult.

An experimentally useful form of Eq. (2) is

$$(\Delta N^2 + 2\Delta N N_T)\delta/I_0 = \delta/R + \alpha\tau_\gamma, \tag{5}$$

where $\delta(T,n) \equiv \Delta(T,n)/\Delta(T,0)$. This factor has been added so that $\alpha = \delta N_T^2/2N_{\omega T}$ becomes independent of T and n for low T. The left-hand side of Eq. (5) can be determined from experimentally measured quantities. Hence, the right-hand side can be determined and the contributions of the two terms can perhaps be separated.

If the δR^{-1} term can be experimentally separated, the n dependence of Δ makes possible the determination of the dependence of R on Δ, information which is otherwise quite difficult to obtain. Theoretical calculations[8,9] yield $R \propto \Delta^m$, with m depending on whether the recombination process involves normal or umklapp phonon emission. For the normal process with the recombining particles at the gap edge, $m = 2$. For the umklapp process there appears to be theoretical disagreement: Rothwarf and Cohen[8] find $m = 1$, while Gray[9] finds $m = 3$. The relative importance of the normal and umklapp processes is also not generally well established experimentally. Theoretically, the umklapp process is found to be about 100 times faster than the normal process in Pb,[8] while in Al they appear to go at comparable rates.[9] Experiments of the type discussed here may be able to resolve some of these questions.

If the $\alpha\tau_\gamma$ term is dominant, the study of R becomes more difficult. In estimating τ_γ, it is important to realize that in a film with two effectively infinite dimensions, phonon random-walk diffusion is important even for $\lambda \gg d$, where λ is the phonon mean free path against pair breaking. This is a typical experimental situation: In Sn, λ is probably ~2000–2500 Å, while a typical film thickness is 500 Å. Assuming that the phonons scatter diffusely at the film surfaces and can leave the film only at the film-substrate interface, we obtain, to a good approximation,

$$\tau_\gamma = \frac{d}{\eta c_s}\left(\frac{\xi(3 + 2\ln\xi)}{2\xi - 1}\right), \quad \xi = \frac{\lambda}{d} > 1. \tag{6}$$

η is the average phonon transmission probability at the film-substrate interface and c_s is the sound velocity. For specular reflection at the film surfaces, and for $\eta = 4\xi^{-1}$ (a condition usually met in practice), we estimate $\tau_\gamma = 4d/\eta c_s$. These two estimates of τ_γ are in essential agreement for typical experimental parameters.

From the above, we see that if $\alpha\tau_\gamma$ is not too large compared with δR^{-1}, it should be possible to separate the two terms using the dependence of τ_γ on film thickness d. The right-hand side of Eq. (5) may display a gap dependence ranging from Δ^{-2} to Δ^0, depending on the relative dominance of various terms and the correct gap dependence of R for umklapp processes. When the τ_γ term dominates, the excess quasiparticle density becomes almost independent of film thickness for a given incident light power. The separation of the various terms of interest, while possible in principle, in practice may place stringent requirements on experimental accuracy.

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†Present address: Institute of Energy Conversion, University of Delaware, Newark, Del. 19711.

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$$dN/dt = I_0 - RN^2 + \beta N_\omega,$$

$$dN/dt = J_0 + \tfrac{1}{2}RN^2 - \tfrac{1}{2}\beta N_\omega - (1/\tau_\gamma)(N_\omega - N_{\omega T}).$$

We can define, in steady state, $\tau_{\rm eff}$ by

$$\Delta N = I_{\rm eff}\tau_{\rm eff},$$

where

$$I_{\rm eff} = I_0 + J_0[2N_T^2 R\tau_\gamma/(2N_{\omega T} + N_T^2 R\tau_\gamma)].$$

One finds

$$N^2 = N_T^2 + I_0[(1/R) + (N_T^2/2N_{\omega T})\tau_\gamma] + J_0[(N_T^2/N_{\omega T})\tau_\gamma]$$

and

$$\tau_{\rm eff} = [(1/R) + (N_T^2/2N_{\omega T})\tau_\gamma](\Delta N + 2N_T)^{-1}.$$

Thus, the formal expression for $\tau_{\rm eff}$ remains unchanged, but in the analysis of data $I_{\rm eff}$ should be used.

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Effects of Dynamic External Pair Breaking in Superconducting Films*

G. A. Sai-Halasz, C. C. Chi, A. Denenstein, and D. N. Langenberg
Department of Physics and Laboratory for Research on the Structure of Matter, University of Pennsylvania, Philadelphia, Pennsylvania 19174
(Received 21 March 1974)

Effects of dynamic external pair breaking in superconducting films are studied using microwave reflectivity to probe excess quasiparticle densities. For weak pair breaking, agreement with theory is good, permitting determination of effective quasiparticle recombination times. For strong pair breaking, an expected first-order transition to the normal state is not observed. Instead, a partially dc-resistive state is found in a broad injection region.

We have experimentally investigated nonequilibrium effects in superconducting Sn films under the influence of external pair breaking, using microwave reflectivity as a probe of the quasiparticle density. The results reported here are interpreted using the theory discussed in the previous Letter.[1] They give new insight into the problem of measuring the intrinsic recombination time of quasiparticles. In addition, they provide a test of the Owen-Scalapino[2] model of a nonequilibrium superconductor under weak and strong pair breaking.

Pair breaking was accomplished with a pulsed GaAs laser ($\lambda = 904$ nm) generating a maximum