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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[7]In general, one can picture both a quasiparticle injection current, I_0, and a flux of phonons with energy greater than 2Δ, J_0. Under these conditions, the rate equations are

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

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

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

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

where

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

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*

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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 peak power of 15 W. cw 70-GHz microwaves reflected from the film were detected and amplified, then averaged in a boxcar integrator and recorded against time. Because of ac coupling, the signal is zero in equilibrium and proportional to $R(0)-R(n)$ (notation follows Ref. 1) in the nonequilibrium situation. Typically, 60–90-nsec pulses were used at a 100-$\sec^{-1}$ rate. The absorbed light energy per pulse was less than 5 ergs at maximum intensity, assuming 62% reflection from the films. This amount of energy was insignificant in raising the substrate temperature; heating effects in general have been discussed earlier.[3,4] On the other hand, the pulses were of sufficient duration to establish quasiequilibrium in the superconductor. The low repetition rate assured the restoration of equilibrium between pulses. The light intensity was variable by a factor more than 10^4. The minimum detectable change in the microwave reflectivity was $2\times10^{-4}\,[R(0)-R_N]$ with an overall system rise time (10–90%) of 25 nsec. The films, ranging in thickness from 150 to 1100 Å, were evaporated on optically flat quartz substrates. Measurements were made in a vacuum can as a precaution against deposition of ice on the film during transfer of helium.

Care was taken to assure uniformity of light intensity over the region of the film probed by the microwaves. The inside of a brass cylinder was fumed with MgO, forming a "white box," the light pipe entered in the side of the cylinder close to the bottom, and the film formed the top side of the cylinder. The probed area was ~25% of the total irradiated portion, placed in the center of it. Over this area the diffusely scattered light was of uniform intensity within our ability to measure it, ~2%.

A typical light pulse (measured by a p-i-n diode photodetector) and a signal are shown in Fig. 1. The top of the signal is flat, indicating that quasiequilibrium is established. The rise and fall times are those of the electronics, implying that the effective quasiparticle recombination time is less than 25 nsec and cannot be measured in this direct way. It can, however, be found indirectly through a measurement of the pulse height (signal amplitude) versus light intensity, as shown in the previous paper. Such data were obtained for all films at several temperatures. The qualitative features of these data were the same in all cases; data points for one case are shown in Fig. 2, together with a theoretical curve derived from Ref. 1. It is apparent that experi-

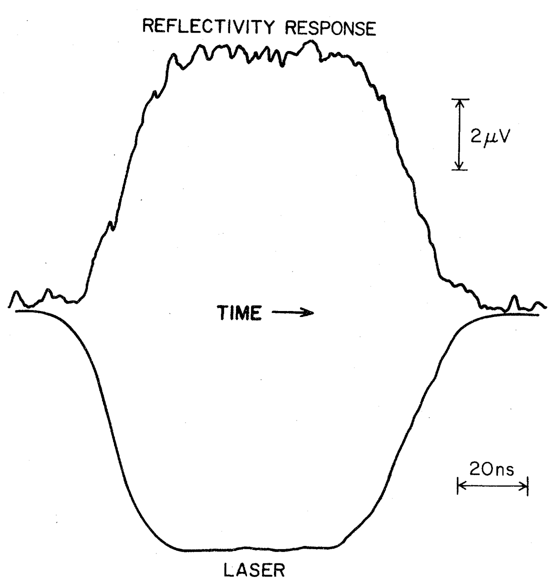

FIG. 1. Typical microwave reflectivity signal and laser pulse. The signal amplitude is proportional to $R(0)-R(n)$, and the voltage scale refers to the microwave detector output. The light is detected by a p-i-n diode, and the scale is arbitrary.

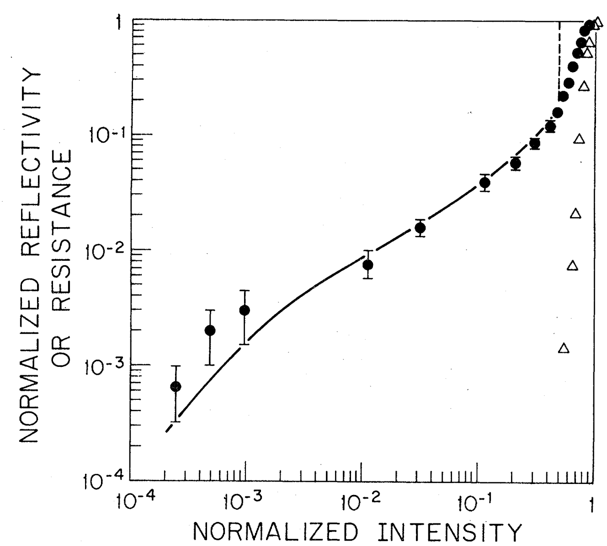

FIG. 2. Microwave reflectivity change and dc resistance as a function of relative pair-breaking intensity for Sn at 1.2 K. Closed circles: reflectivity change $R(0)-R(I)$ normalized with respect to the full change $R(0)-R_N$ between the equilibrium superconducting and normal states for a 400-Å-thick film; intensity at 1 is 8 W cm^{-2}. Solid line: theoretical curve of Ref. 1. Dashed line: where the predicted first-order transition should have occurred. Triangles: resistance $r(I)$ normalized to the normal-state value r_N for a 500-Å strip; for this curve, absolute light power at 1 is 7.6 W cm^{-2}.

ment and theory are in good agreement over 3 orders of magnitude in light intensity and signal. (The figure shows the least satisfactory fit of all the cases studied.) However, the sharp transition to the normal state at high light intensities predicted by the Owen-Scalapino modes does not occur! We shall return to this feature below, after considering the implications of the region where theory and experiment do agree.

In all cases the theoretical curve was fitted to the data by adjusting only the light-intensity scale, using the relation $CI_0 = \Delta N^2 + 2N_T\Delta N$. The scaling constant C contains the physically interesting parameters. The importance of τ_γ, the time required for recombination phonons to leave the film, can now be determined. In this event, $C = 1/R + N_T^2\tau_\gamma/2N_{\omega T}$ should display a dependence on film thickness. The data plotted in Fig. 3 show that this is indeed the case. In comparing values of τ_γ obtained from these data with the theoretical estimates of Ref. 1, it should be borne in mind that, in addition to other limitations of the latter estimates, the wavelength of the recombination phonons is more than 100 Å, approaching the thickness of the films used. This undoubtedly further limits the reliability of the theoretical estimates. However, the experimental values of τ_γ are of the right magnitude. For our 900-Å film (where the nonzero wavelength of the recombination phonons should have the least effect), assuming specular reflection of phonons from the film surfaces (the diffuse reflection case is not very different), we find the phonon escape probability $\eta \simeq 7\times10^{-2}$, taking the conversion efficiency of the light power $F = \frac{3}{4}$. This is in reasonable agreement with values estimated by combining experimental Kapitza resistance measurements[5] and theory.[6]

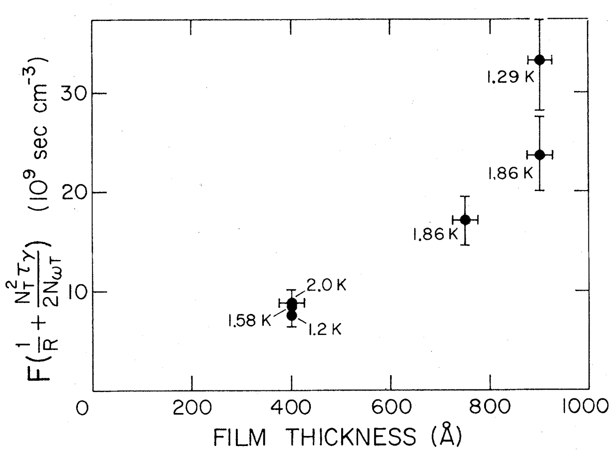

FIG. 3. Fitting parameter as a function of film thickness. This parameter is obtained from reflectivity data, as in Fig. 2. Error bars represent relative errors; an overall 30% uncertainty exists due to the uncertainty in absolute light power.

In principle, one may attempt to obtain the intrinsic[7] recombination time $\tau_R = (2N_TR)^{-1}$ either from extrapolation of data like that in Fig. 3 to zero film thickness, or directly from measurements on very thin films. In one attempt on a 150-Å film, we have established the existence of effective quasiparticle lifetimes much shorter than in the thicker films, and we are currently pursuing this possibility. The extrapolation procedure imposes severe requirements on experimental accuracy; it is apparent that the data of Fig. 3 permit only the establishment of an upper limit on τ_R. Assuming that τ_R is not much larger than the uncertainty (~25%) in $\tau_{\text{eff}}(\Delta N = 0)$ for the 400-Å film (where the τ_γ term still clearly dominates), we find $\tau_R \leqslant 3\times10^{-11}F^{-1}t^{-1/2}\exp[\Delta(T)/kT]$. To our knowledge, this is the first time that the overwhelming contribution of τ_γ in the quasiparticle recombination process has been established, and a limit given for the intrinsic recombination time.

Returning now to the absence of the expected first-order transition to the normal state, we note (cf. Fig. 2) that, although the slope of the signal-amplitude versus light-intensity curve does increase at high light intensity, the reflectivity approaches its normal value over about a factor of 2 in light intensity, converging to it with essentially zero slope. Limiting the film sizes to the area of microwave incidence had no effect, indicating that quasiparticle diffusion out of the probed area was of no importance.

To gain more insight into the state of the superconductor in the region near the transition, we correlated dc measurements with the microwave measurements. We used strip films, typically 1.5 mm wide and 2 cm long. The microwaves and light were incident on the middle on a spot of ~0.5 cm diam. Outside the illuminated area, leads for a four-terminal resistivity measurement were connected. Measuring currents were typically 1–5 mA. The electronic processing of the dc voltage signals was the same as for the microwave signals. Signals stemming from an Ohmic resistance (inductive effects are insignificant) appear, with zero slope, close to the point where the transition should have occurred, and gradually increase with pair-breaking intensity. The shapes of the dc-signal pulses are exact replicas of the microwave signal pulses. The maximum dc signal corresponded exactly to the normal resistance of the film strip, as measured the standard way at 4.2 K, and in light intensity corresponded to the point where the microwave reflectivity indicated the normal state. For a given light intensity the dc signals displayed a perfectly Ohmic character. The dc current had no effect on the microwave signals and vice versa. Typical dc data points are shown in Fig. 2.

It appears from both the microwave and dc measurements that, rather than undergoing a sharp transition to the normal state, the films undergo a transition to what may be a new type of superconducting state, a sort of photon-induced dynamic intermediate state. Following is a highly speculative discussion which, in our view, represents a possible picture of this state. It is known that the recombination phonons play a central role in the recombination dynamics. Suppose that a critical excess quasiparticle density is reached and the film turns normal discontinuously. In the normal state, there is no bottleneck for the incoming energy flux in the form of quasiparticles of energy $\sim\Delta$ and phonons of energy $\sim 2\Delta$, that are the most effective in impeding pairing. The film goes back to the superconducting state, starting the process over. The larger the pair-breaking intensity, the shorter the time the film spends in the superconducting state. Beyond the temporal fluctuations, spatial breakup is probably inevitable, but of secondary importance, due, for example, to slight inhomogeneities in film thickness. If the temporal scale is $\lesssim 10$ nsec, both the microwave and dc signals would indicate an average of the measured parameter, behaving as if a well-defined fraction of the film is normal at all times.

We have been able to put an upper limit on the time scale of these processes. The dc signals at sufficiently high intensities are large enough to be observed directly on a sampling oscilloscope. The measured fall time was 1 nsec. However, this might have resulted from the fall time of the light intensity and must be regarded as an upper limit.

A proper theoretical understanding of this dynamic intermediate state will obviously require consideration of a possible superconducting-normal interface energy, and of the bremsstrahlung phonons created in the cooling of the primary high-energy particles.

Another feature of our data should be mentioned: The change in the microwave reflectivity of the films is delayed measurably after arrival of the light. This "initial delay" was proportional to thickness (approximately 1 nsec per 100 Å), but independent of the light intensity. The latter indicates that the initial delay is not connected with the recombination process. In most of our experiments, the light was incident on the side of the film opposite to that which the microwaves sampled, and the possibility arises that it takes this delay time for the effect to cross the film. This in turn could be an indication of a nonuniformity across the film, making our results questionable. However, both quasiparticles and phonons cross the films in less than 10^{-10} sec, making it virtually impossible for nonuniformity to exist on the 10^{-8}-sec time scale of our work. To check this point, in one case we irradiated a film from the front side. No differences were noted in any respect; in particular, the initial delay was unchanged. This leads us to believe that the initial delay is the result of the cascade process through which each photon distributes its energy among quasiparticles and phonons. It is known[8,9] that high-energy electrons are dumped overwhelmingly by phonon emission, rather than by pair breaking. Accordingly, the cascade process involves several phonon emission and adsorption steps, the total time of which can be of the order of a few nanoseconds. However, we measured the initial delay also in a Pb film where, according to the same theory, the emission and adsorption processes are at least an order of magnitude faster, and found approximately the same initial delay as in similar thickness Sn films. This leaves us with no firm explanation of the phenomenon, or its thickness dependence.

Shortly before submitting this Letter it came to our attention[10] that photoinduced effects in superconductors have also been investigated independently by other groups.

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Liquid-Gas Phase Diagram of an Electron-Hole Fluid

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Measurements and theoretical estimates are presented of the liquid-gas phase diagram of an electron-hole fluid in pure germanium. The critical point is found to occur in a region where the fluid is an electron-hole plasma at $T_c = 6.5 \pm 0.1$ K and $n_c = (8 \pm 2) \times 10^{16}$ cm^{-3}.

We present here a study of the phase diagram of the electron-hole condensate in germanium, based upon information obtained from the luminescence spectrum under constant optical pumping. The luminescence from the condensed phase, which has been observed in Si[1] and Ge,[2] indicates (from the low-temperature line shape[2-4] and its temperature dependence[4]) that the condensate is an electron-hole liquid. While it is well established that excitons exist at low carrier densities in the gas phase,[5] we shall present evidence that the gas is an electron-hole plasma at high densities near the critical point. We shall discuss only briefly the transformation between excitons and plasma, since it is not as yet understood.

The liquid-gas phase diagram for electrons and holes in germanium has been measured using two different aspects of the recombination luminescence. First, the line shape of the luminescence from the liquid has been used to obtain its absolute density as a function of temperature. This procedure has been discussed previously[4] for temperatures below 4.25 K and is here extended to higher T. Second, onsets of liquid luminescence as a function of T at constant pumping power have been used to determine the relative density in the gas at the boundary of the two-phase region. Onsets analogous to these have been measured in luminescence,[2,6] in cyclotron resonance,[7] and in noise pulses,[8] in the temperature region below 4.2 K.

At high densities the electron-hole liquid is metallic and there is good agreement between the first-principles calculations,[9-11] based on the random-phase approximation (RPA) at $T = 0$ and modifications thereof, and experiment. As temperature is raised the density n of the liquid is reduced through thermal expansion.[4] Within the RPA one may write the free energy per particle as

$$F(n,T) = F_0(n,T) + F_{xc}(n,T), \qquad (1)$$

where F_0 is the free energy of a noninteracting set of electrons and holes and F_{xc} represents the exchange and correlation corrections due to the interactions. In the metallic regime one can, to a good approximation, ignore the explicit T dependence of F_{xc} and write $F_{xc}(n,T) \equiv F_{xc}(n)$. The validity of this approximation, which we have checked by direct calculation, can be understood since, within the RPA, the excitation spectrum is strongly perturbed by the interactions only at energies of order of the plasma frequency ω_p. Since, in Ge, $\omega_p \approx 14\tilde{n}^{1/2}$ meV, where $\tilde{n} = n \times (10^{-17}$ cm$^3)$, the condition $k_B T \ll h\omega_p$ is satisfied. The main effect of the interactions is to lower rigidly the bottom of the band with only a negligible change in effective mass.[12] This effect diminishes rapidly as the density drops. It can be estimated theoretically by computing the exchange and correlation contributions to the chemical potential, $\mu_{xc}(n) \equiv F_{xc} + n\partial F_{xc}/\partial n$. Finally, on theoretical grounds, one might expect that at a sufficiently low density $n = n_{MI}$ the fluid ceases to be metallic, where

$$n_{MI} = (1/8\pi)(k_B T/E_x)a_x^{-3}. \qquad (2)$$

This is the Mott criterion for nondegenerate carriers (based on the Debye screening length) where E_x and a_x are the exciton Rydberg and Bohr ra-