Pa.; Model No. L0904-125-H-T2.

[15]The magnetoresistance of the thermistor was measured at the boiling points of liquid nitrogen and liquid argon, with the thermistor in each case immersed in a short column of the boiling liquid. The magnetoresistance near T_N was then obtained by interpolating between the two sets of data. Additional measurements of the magnetoresistance near T_N were made with the same arrangement (with the two cans) as in the thermal expansion measurements. Each data point near T_N was taken by first stabilizing the temperature and then changing H from zero to a given value and then back to zero. The time in which the field was changed was short compared to the time in which the temperature drifted by several millidegrees Kelvin, but was sufficiently long to avoid significant eddy-current heating. The results of the two methods of determining the magnetoresistance near T_N were in agreement with each other.

[16]Y. Shapira and S. Foner, Phys. Rev. B 1, 3083 (1970).

Quasiparticle Propagation and Recombination in Bulk, Superconducting Pb

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The propagation characteristics of quasiparticles in bulk superconducting Pb single crystals is studied. A transition from quasiparticle diffusion to diffusion of the combined gas of quasiparticles and phonons is observed as the temperature is increased. The *intrinsic* quasiparticle recombination time, as well as the decay time of the quasiparticle density, is determined. The latter is found to be at least an order of magnitude longer than this quasiparticle recombination time.

Recently there has been considerable activity in the study of nonequilibrium superconductivity.[1] Most of the experimental studies to date have involved a study of the superconducting transition of a thin film or the *I-V* characteristics of thin-film tunnel junctions under the influence of laser excitation. In this Letter, we report on the propagation characteristics of photoexcited quasiparticles in bulk, single-crystal, superconducting lead using a tunnel junction as a quasiparticle detector and time-of-flight techniques with nanosecond resolution. The combined use of *bulk* samples and time-of-flight techniques makes possible a *direct* determination of the spatial and temporal behavior in the nonequilibrium state in a heretofore unexplored regime.

We present data which show a transition from diffusive heat propagation (in the combined gases of quasiparticles and phonons) to a pulse of diffusing quasiparticles as the temperature is lowered below $T \sim 2.8$ K. The quasiparticle scattering rate, in the high-temperature regime, is shown experimentally to be equal to the quasiparticle recombination time as determined from calculations using tunneling data. The low-temperature pulse reflects the decoupling between the local temperature and the quasiparticle number.

The experiments were performed on high-purity (99.9999%) Pb single crystals with propagation direction [111] and thicknesses 4.5, 2.2, and 0.87 mm. The resistance ratio ($R_{300\text{ K}}/R_{2\text{ K}}$) of these samples is of the order of 20 000. The samples were polished using a combined mechanical and chemical technique and suspended from a single point to avoid straining them during the cooldown in the liquid helium cryostat. One surface of the sample was cleaned by back-sputtering in argon to remove undesirable surface contaminants. A controlled oxide layer was then grown on this surface and a thin lead film was evaporated on top to form the tunnel junction. The size of the junction was $\sim 0.25 \times 0.45$ mm^2 and the normal-state resistance was $\sim 20-50$ mΩ. Silicon oxide and photoresist were used to insulate the lead film from the lead crystal outside the junction area. The photoexcited quasiparticles were generated at the opposite surface of the crystal by means of a dye laser which was in turn pumped by a nitrogen laser. The laser had a peak power of ~ 1 kW (only a fraction of which was absorbed by the crystal) with a rise time of 1.5 ns and a pulse width of 5 ns. The beam was about 0.2 mm in diameter. The laser and power supply were located outside a shielded room in which the cryostat was placed to eliminate undesirable electrical pickup. Great care was also taken to eliminate scattered light from reaching the detector. The voltage signals from the tunnel detector (biased in the region of thermal quasiparticle tunneling) were amplified by means of a B & H amplifier with 2-GHz bandwidth. In the initial experiments a PAR 162 boxcar integrator with a 400-ps gate

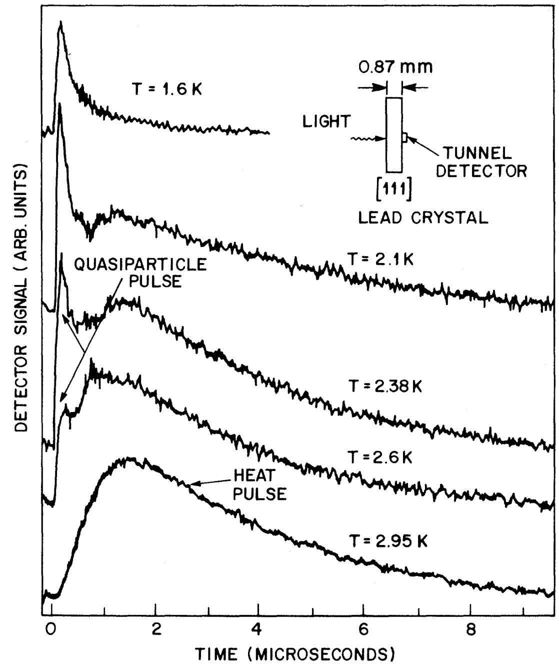

FIG. 1. Tunnel detector signal as a function of time for five different temperatures. Single-crystal lead; propagation length 0.87 mm; orientation [111]. As the temperature is lowered below about 2.6 K one observes the decoupling of the quasiparticles from the heat pulse. See text.

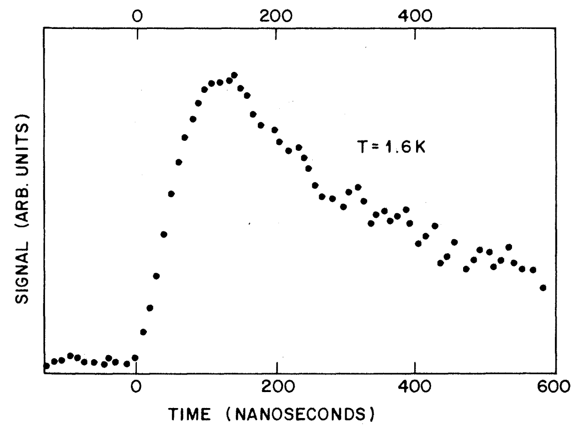

FIG. 2. Expanded trace of the low-temperature (1.6 K) quasiparticle pulse. Propagation length and orientation same as in Fig. 1.

was used for signal averaging. Because the arrival time of the signal was later than originally expected, later measurements (and the ones presented below) were taken with a Biomation 8100 transient recorder with 10-ns resolution and a Nicolet signal averager for considerably improved signal-to-noise performance.

In Fig. 1, we show some typical pulses detected by the tunnel junction in the temperature range of 1.6 to 2.95 K for the 0.87-mm crystal. In this figure we have also sketched the geometry of the experiment for clarity. At $T = 2.95$ K and higher, only a single broad pulse with a shape characteristic of a diffusive heat pulse is detected. As the temperature is lowered below about 2.8 K, a new faster pulse splits off from the heat pulse and grows in intensity. Below about $T \approx 2$ K only the faster pulse remains. Its leading edge has a rise time of somewhat less than 100 ns and an exponentially decaying tail. An expanded trace (in time space) of this low-temperature pulse is shown in Fig. 2. The apparent similarity of this pulse shape with the 2.95-K diffusive pulse in Fig. 1 leads us to believe that this quasiparticle pulse is diffusive. However, the peak arrival time does not increase as the square of the propagation length L (as is observed in the higher-temperature regime). Rather it shows an almost linear L dependence. It is also important to point out here that the detected pulse shapes are not limited by the time constant of the tunnel-junction detector. This was verified by shining light directly on the junction.

We now turn to possible physical models for the above-mentioned data, considering first the data in the region from 1 to 2 K. The exciting light pulse serves to generate a "hot" gas of quasiparticles excited to high energies. These particles relax very rapidly to within a few kT of the gap edge.[1] The scattering rate of these quasiparticles is then limited by four different processes: (a) quasiparticle-quasiparticle Coulomb scattering time, τ_{qq}; (b) quasiparticle-impurity scattering time, τ_i; (c) quasiparticle-phonon scattering time τ_S; and (d) quasiparticle-quasiparticle recombination, τ_R, to the condensate via phonon emission. From theoretical computations of Kaplan *et al.*[2] we may expect τ_S and τ_R to become longer than the quasiparticle ballistic propagation time[3] ($\tau_B \sim 10^{-8}$ sec) at temperatures of 1–2 K. Although τ_{qq} has not been measured in Pb, reasonable theoretical estimates give values of order 10^{-5} sec, much too long to be important in present experiments. τ_i is, however, shorter than τ_B and is estimated to be $\sim 2\times10^{-10}$ sec for a resistance ratio of 20 000. This implies that the quasiparticles should travel by impurity-scattering–limited diffusion below 2 K. For a *point* source in an *infinite* medium, the quasiparticle density $\Delta n(r, t)$ at a distance r and time

t for a δ-function input pulse should have the diffusive form

$$\Delta n(r, t) \propto t^{-3/2} \exp(-r^2/4Dt), \tag{1}$$

where $\boldsymbol{D}$ is the diffusion constant of the quasiparticles given by

$$D_{\mathrm{qp}} = \tfrac{1}{3}\langle v^2 \rangle \tau_i. \tag{2}$$

Here $\langle v^2 \rangle$ is the mean thermal quasiparticle velocity. D can be obtained[4] from (1) by taking the tangent at the steepest point on the leading edge of the pulse. The point of intersection of this tangent with the time axis occurs at a time t given by

$$t = 0.0375 L^2/D. \tag{3}$$

Using (2) and Fig. 2, we estimate τ_i to be $\sim 2 \times 10^{-10}$ sec in excellent agreement with the resistance-ratio measurement supporting our contention that this pulse is impurity-limited diffusion.

We believe that the anomalous length dependence is related to the exponential nature of the trailing edge of the pulse. Under the assumption of an $\exp(-t/\tau_{\mathrm{damp}})$ damping process modulating the line shape of Eq. (1), in the limit $4r^2/4\boldsymbol{D}\tau_{\mathrm{damp}} > 1$, the peak arrival time, t_{peak}, scales approximately with length. For damping times shorter than 10^{-7} sec, this condition is satisfied. Measurements of the temperature dependence (in the region 1 to 2 K) of this exponentially decaying tail and possible physical interpretations will be published in a detailed paper.[5]

From broad-band bolometer data[5] between 1 and 2 K we observe low-frequency ballistic phonons arriving at ≈ 1 μsec. These, of course, are not detected by the tunnel junction. Thus, in reality, at low temperatures there are two decoupled pulses propagating through the crystal; one predominantly quasiparticle in nature, the other phonon in nature. On raising the temperature above 2 K, we expect τ_S and τ_R to be comparable and $\sim 5 \times 10^{-9}$ sec. The onset of this cross-coupling between the quasiparticles and the phonons results in the tunnel junction detecting two pulses between 2 and 3 K. Above 3 K the rapid thermalization yields only a single heat pulse which evolves from this pair of pulses. This single higher-temperature heat pulse is detected with either the bolometer or the tunnel junction.

Using Eq. (1) and the usual kinetic model for heat flow, we can write the diffusion constant, D_{qp}, as

$$D_{\mathrm{qp}} = \tfrac{1}{3}(C_{\mathrm{qp}}/C_{\mathrm{tot}})\langle v^2 \rangle \tau_R/2. \tag{4}$$

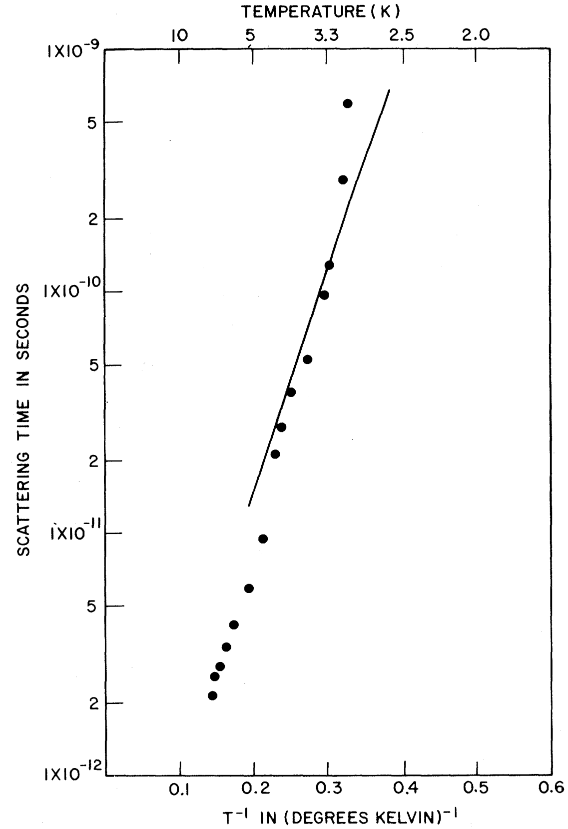

FIG. 3. Experimental quasiparticle scattering time as a function of T^{-1} (solid circles). The solid line is the theoretical recombination lifetime at $\Delta(T)$ obtained from Ref. 2.

Here C_{qp} and C_{tot} are the quasiparticle and total specific heats, respectively, and the dominance of τ_R over τ_i and τ_S is assumed. The factor of 2 in Eq. (4) occurs because of the two-particle nature of the recombination process. The results of such an analysis are shown in Fig. 3 as the solid circles. Also shown (solid line) is a theoretical curve for the recombination lifetime at $\Delta(T)$ taken from the paper of Kaplan *et al.*[2] The theoretical curve is shown only for temperatures below about 5 K. Above this temperature, kT is significant compared to $\Delta(T)$ so that recombination lifetime is effectively shorter than at $\Delta(T)$ in agreement with experiment. The numerical agreement between the two curves is to be considered excellent and lends strong support to the interpretation given here.

Having shown that the observed transition can be interpreted in terms of a transition from a heat pulse to a pure quasiparticle pulse, one must ask how and what aspects of the quasiparticles no longer relax rapidly to equilibrium with the acoustic phonons. We find from various estimates that it is the quasiparticle *number* that is out of equilibrium. The interesting feature of these data is that the relaxation time of the quasiparticle number must be greater than the arrival time of the quasiparticle pulse. This implies quasiparticle-number decay times of >100 nsec. This is much longer than the quasiparticle recombination rate τ_R. The fact is that the 2Δ phonon created by the recombination of two quasiparticles lives for a very short time, almost immediately decaying back into two quasiparticles. It does occasionally decay into two phonons of energy less than 2Δ so that the effective quasiparticle number lifetime is $(\tau_R/\tau_{p\to 2q})\tau_{p\to 2p}$. Using a value of 10^{-9} sec for $\tau_{p\to 2p}$ obtained for phonons of energy 2Δ from the work of Narayanamurti, Dynes, and Andres[6] on Bi and a value of $\tau_{p\to 2q} \sim 3\times 10^{-11}$ sec,[2] we obtain a ratio of $\tau_{p\to 2p}/\tau_{p\to 2q} \sim 30$ in rough agreement with our observations.

Using these concepts we have developed a coupled set of diffusion equations to describe the transition. The two equations couple the local temperature and the quasiparticle number. At weak coupling, the two equations predict *two* diffusive pulses. With increased coupling, the two pulses merge into a single diffusive pulse as is observed experimentally. A description of the theory will be given in a full report of the work.[5]

In summary, we have studied the spatial and temporal behavior of photoexcited quasiparticles in bulk single crystals of Pb using a tunnel junction as a quasiparticle detector. At low temperatures, a quasiparticle diffusive pulse limited by impurity scattering is observed. Above 3 K the observed pulse is in the combined phonon and quasiparticle gases and an analysis allows a direct determination of the intrinsic quasiparticle recombination time.

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[1]For a recent review see D. N. Langenberg, in *Proceedings of the Fourteenth International Conference on Low Temperature Physics, Otaniemi, Finland, 1975*, edited by M. Krusius and M. Vuorio (North-Holland, Amsterdam, 1975).

[2]S. B. Kaplan, C. C. Chi, D. N. Langenberg, J. J. Chang, S. Jafarey, and D. J. Scalapino, Bull. Am. Phys. Soc. 21, 258 (1976), and to be published. We would also like to thank S. Jafarey and D. J. Scalapino for sending us computed numerical values for τ_S and τ_R for Pb.

[3]This was calculated from the known Fermi velocity [see G. I. Lykken, A. L. Geiger, K. S. Dy, and E. W. Mitchell, Phys. Rev. B 4, 1523 (1971)] and the assumption of a BCS superconductor.

[4]See, for example, C. C. Ackerman and R. A. Guyer, Ann. Phys. (N.Y.) 50, 128 (1968).

[5]P. Hu, R. C. Dynes, V. Narayanamurti, H. Smith, and W. F. Brinkman, to be published.

[6]V. Narayanamurti, R. C. Dynes, and K. Andres, Phys. Rev. B 11, 2500 (1975).