Direct Real Time Measurement of Quasiparticle Lifetimes in a Superconductor

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A short laser pulse breaks Cooper pairs and photoexcites electrons in a superconducting film of niobium. As the quasiparticles recombine and rejoin the condensate, changes in the electrodynamic response of the sample are detected by a fast oscilloscope. The observed biexponential decay is consistent with a previous measurement. The fast recovery time is used to obtain the pair recombination time, which is in good agreement with theory.

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Many of the fundamental properties of superconductors are intrinsically nonequilibrium in nature. In particular, the rate at which single-particle excitations (quasiparticles) recombine into pair states and rejoin the condensate is a measure of the coupling between electrons and phonons in a BCS superconductor. Despite great effort, there are few experimental techniques available to measure quasiparticle lifetimes, and most of these are indirect. The more common techniques [1] have used double-junction (S-I-S-I-S or N-I-S-I-N, where S is a superconductor, I an insulator, and N a normal metal) sandwich geometries. One junction is used as a source of injected quasiparticles and the other detects their density which, in a steady-state process, is proportional to the product of the injection rate and the lifetime. In the latter geometry [2], the electron branch of the quasiparticle spectrum is overpopulated with respect to the hole branch, and a "branch imbalance" is detected as an excess voltage by the second, normal-metal electrode. A variation of this method [3] measures the diffusion length of a branch imbalance created in a quasi-one-dimensional wire. Because quasiparticle lifetimes are short (typically of order 1 nsec) and the excess voltages are small, these techniques have only been applied to a few materials, such as Al and Sn, which have relatively long lifetimes. In a seminal experiment [4] that was much more direct, the order-parameter relaxation time near the transition temperature T_c was deduced by subjecting a sample to a large dc bias current and a small rf current of variable frequency, and measuring a frequency-dependent rf voltage caused by the kinetic inductance of the superconducting carriers.

The photoresponse of superconductors was first directly studied by Testardi [5], who measured the resistance of thin superconducting Pb films during illumination by pulses of 514-nm laser light. He demonstrated that the response below T_c could not be simply explained by a model in which the film is heated above T_c during the pulse. Other experiments [6] studied tunnel junctions, or measured microwave reflection from illuminated surfaces. In this Letter we apply an improved measurement of the photoresponse to *directly* measure the quasiparticle recombination time τ_r from voltage transients in a superconducting sample that has been photoexcited by short pulses of laser light. Our experiment uses a very fast transient oscilloscope and a geometry optimized to eliminate flux flow due to edge effects. Times are measured for Nb in the range of reduced temperature T/T_c from 0.75 to 0.95; τ_r is so short in Nb that it has previously been measured only once [7]. That experiment used a branch-imbalance diffusion-length measurement and unexpectedly observed [8] a biexponential decay. The present measurement confirms that novel result. Furthermore, the fast decay time of the biexponential is used to obtain a value for τ_r which is in good agreement with the theoretically predicted value [9].

The apparatus [10] is depicted in Fig. 1. A thin Nb film of thickness d (20 or 40 nm) is sputtered on a transparent substrate. A 200-nm-thick gold film is deposited over an annular mechanical mask leaving uncoated a portion of the Nb film, with an inner diameter of 0.5 mm and an outer diameter of 1.5 mm, which terminates a 50-Ω coaxial transmission line [Fig. 1(a)]. A bias tee allows the sample to be biased with a dc current I_b, and a Hypres psec signal processor [11] (with temporal resolution better than 15 psec) is connected to the other end of the transmission line [Figs. 1(b) and 1(c)]. With I_b flowing radially in this (Corbino disk) geometry, magnetic-field lines are parallel to the plane of the sample, and contributions to the response due to vortex motion at edges or at grain boundaries, as can occur in linear geometries, are minimized. The sample is illuminated through the substrate by 300-fsec pulses of 665-nm light at a repetition rate of 2 kHz [12] with a beam diameter of 1.8 ± 0.2 mm. The sample is heat sunk to a copper block fixed to, but electrically isolated from, the cold finger of a gas-flow cryostat. Diode thermometers measured changes in temperature with an accuracy of better than 0.01 K. The transition temperatures of the films were determined independently.

The response of a superconducting film to pulsed laser light is in general highly nonlinear, and is linear only when a low fluence causes a weak perturbation [13] such that the resulting number of nonequilibrium quasiparticles δN_{qp} is small compared with the thermal-equilibrium number N_{qp}. In this regime the response can be conceptualized as follows. The optical penetration depth of Nb is about 10 nm, and the sample absorbs about 40% of the

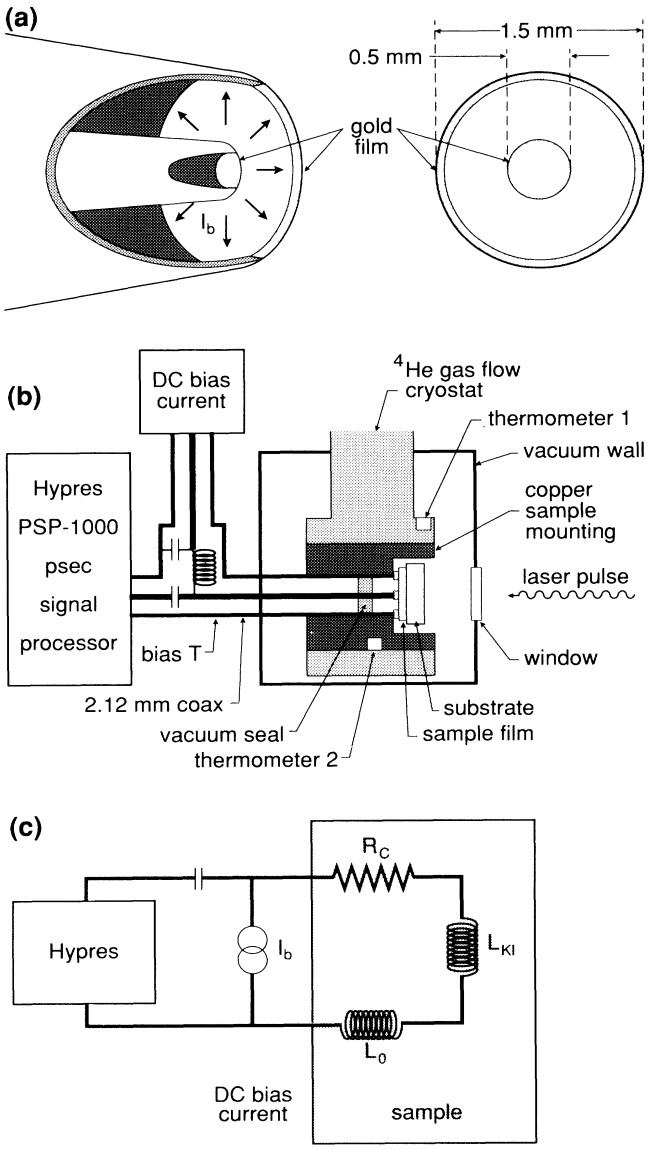

FIG. 1. (a) Sample geometry. The superconducting film terminates a 50-Ω rigid coaxial cable (shown cut away) and is biased with a dc current from center to outer conductors. End view at right. (b) Block diagram of the experimental configuration. (c) Simplified circuit schematic for $T < T_c$. R_c is a contact resistance, L_0 is a stray inductance, and L_{KI} is the kinetic inductance of the superconducting channel of the sample. The resistive impedance of the normal channel which shunts L_{KI} (in a two-fluid model) is not shown.

incident energy [14]. The absorbed photons break pairs and excite electrons to an energy $E' = 1.9$ eV above the Fermi level E_F. In less than a psec, five or six generations of electron-electron collisions occur such that each of the original photoexcited electrons produces a number of electrons (high-energy quasiparticles) with energy comparable with the Debye energy. Relaxation proceeds [13] either by electron-electron scattering or by phonon emission (with possible subsequent pair breaking by the phonon before it leaves the film). The time scale for these processes in Nb is of the order 10 psec [13]; after three to five generations the quasiparticles have mean excitation energy near the gap, $E' \sim \Delta$, and recombination dominates relaxation events for $E' < 2\Delta$ [15]. Thus, after a time $t_0 \sim 50$ psec there is a distribution of quasiparticles that is believed to differ only slightly from thermal equilibrium. These quasiparticles rejoin the condensate by emitting a phonon of energy $\hbar\Omega \approx 2\Delta$. This phonon may break a pair and form two new quasiparticles if the pair-breaking time τ_B is short compared to the time τ_γ for it to become unavailable for pair breaking. This time is a combination of the time to escape from the sample τ_{es} and the time τ_s^{ph} to scatter into lower-energy phonons that are unable to break pairs, $\tau_\gamma^{-1} = (\tau_{es})^{-1} + (\tau_s^{ph})^{-1}$.

The interaction between pairs, quasiparticles, and phonons is governed by the Rothwarf-Taylor coupled equations [16]. In the weak perturbation limit they can be linearized [17]:

$$\frac{d\delta N_{qp}}{dt} = -\frac{\delta N_{qp}}{\tau_r} + \frac{2\delta N_{ph>}}{\tau_B},$$
$$\frac{d\delta N_{ph>}}{dt} = \frac{\delta N_{qp}}{2\tau_r} - \frac{\delta N_{ph>}}{\tau_B} - \frac{\delta N_{ph>}}{\tau_\gamma}, \quad (1)$$

where δN_{qp} and $\delta N_{ph>}$ are the energy-averaged densities of excess quasiparticles and phonons with energies greater than the gap, and τ_r is the average time for two quasiparticles to recombine as a pair. For the time-dependent solution [17] appropriate to a pulsed experiment, the quasiparticles recombine exponentially with a lifetime τ_{eff} that is enhanced over the intrinsic recombination time τ_r by the phonon trapping factor, $\tau_{eff} = \tau_\gamma + \tau_r(1 + \tau_\gamma/\tau_B)$. For larger perturbations the Rothwarf-Taylor equations are nonlinear and the time-dependent problem is not analytically solvable [13]. A useful approximation [15] is to describe the nonequilibrium phonon distribution by a thermal distribution at an effective temperature T^*. Our data at high fluences are consistent with a T^* model [18], which gives additional confidence to the interpretation of low-fluence data presented herein.

The relationship between the relaxation of a nonequilibrium distribution of quasiparticles and the electrical response of a superconductor under fast-optical-pulse excitation is not clear at present. A simple kinetic-inductance model, where the transient voltage is given by $V_{sig} = I_b\, dL_{KI}/dt$, suggests a dynamic response under dc bias corresponding to a decreasing inductance as the excited quasiparticles recombine. This would lead to a negative signal voltage. However, the observed signal is positive and has magnitude larger than expected for a kinetic inductance, suggesting a basically resistive response. In light of the high-frequency nature of the dynamics, a more complete high-frequency model (including the normal channel, such as in the work of Chang and Scalapino

[19], who have calculated the rf impedance of a superconductor under nonequilibrium conditions) may be required to describe properly the photoresponse of a superconductor.

Four Nb samples with thicknesses $d < \lambda$, where λ is the London penetration depth, were studied using I_b of 1–300 mA and laser fluences of 3 nJ/pulse to 3 μJ/pulse. The response had an amplitude that was linear with I_b for values $I_b < I_c/3$, where $I_c(T)$ is the critical current, and a decay time that was independent of I_b in this range. Both amplitude and decay time were highly nonlinear as a function of laser fluence for all but the lowest fluences [18]. We restrict our attention to the linear regime (typically < 5 nJ/pulse) such that a small change in fluence caused a proportionate change in amplitude, but no change in decay time. A semilogarithmic plot of a typical signal is shown in Fig. 2 for time $t > t_0$. The signal amplitude corresponds to an impedance $Z_{sig} \approx 30$ mΩ, i.e., about $Z_{sig}/R_N \approx 0.3$, where R_N is the sample resistance in the normal state. This represents a response, scaled to the incident optical energy per pulse, of 1×10^7 Ω/J. All samples showed null response for no illumination; base lines determined under control conditions were subtracted from data sets before analysis. Above T_c all samples showed a signal with a response of about 2×10^4 Ω/J [20], i.e, several hundred times smaller.

For the low-fluence, low-bias-current conditions of the data in Fig. 2, we estimate $\delta N_{qp}/N_{qp} < 10^{-1}$, consistent with the assumptions underlying Eqs. (1) (we also note that $t_0 \approx 100$ psec as expected). The signal is well fitted by a biexponential decay with a fast time τ_{eff} of 230 psec and a slow time τ_2, discussed below, of 2.6 nsec. Some typical values of τ_{eff} and τ_2, taken from thirty (low-fluence) data sets on three samples, are given in Table I and illustrate their dependence on temperature and on film quality. Variations of the values from sample to sample can be correlated with film quality. Sample 1 had

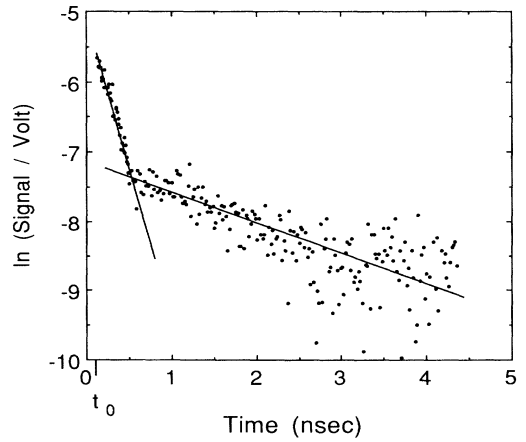

FIG. 2. Semilogarithmic plot of signal voltage vs time for sample 1 at $T/T_c = 0.78$, and fits. Incident fluence is 2.8 nJ/pulse.

TABLE I. Representative values of τ_{eff} and τ_2 for three samples: sample 1, 400-Å Nb on sapphire; sample 2, 200-Å Nb on sapphire; sample 3, 400-Å Nb on lanthanum aluminate. Some blank spaces in the table result because fits for τ_{eff} and τ_2 at a given temperature may be from different data sets (short- and long-time scales, respectively). Errors refer to the fits. Multiple values at the same temperature indicate reproducibility of results.

Sample number	τ_{eff} (psec)	τ_2 (nsec)	T/T_c	T_c (K)
1	220 ± 20		0.75	9.2
1	230 ± 30	2.60 ± 0.30	0.78	9.2
1	240 ± 20		0.78	9.2
1	580 ± 20		0.95	9.2
2	430 ± 20	3.26 ± 0.30	0.76	8.8
2	430 ± 20	6.0 ± 0.30	0.76	8.8
3	650 ± 150	2.81 ± 0.30	0.81	8.7
3	390 ± 100	3.68 ± 0.30	0.81	8.7
3	560 ± 100	7.14 ± 0.40	0.92	8.7
3	980 ± 200	10.31 ± 0.40	0.92	8.7
3	1070 ± 200		0.93	8.7

the highest value of T_c (9.2 K), followed by sample 2 (8.8 K) and sample 3 (8.7 K). Niobium readily forms a suboxide [21], and small amounts of oxygen can degrade a sample and depress T_c. Values of τ_{eff} are longer for the lower-quality samples; a fourth sample with a T_c of 8.4 showed even longer decay times. Because the T_c of sample 1 is the same as bulk Nb, its values of τ_{eff} can be related to τ_r for pure Nb. The phonon escape time τ_{es} can be estimated as [22] $\tau_{es} = 4d/\eta c_s$, where d/c_s is the time it takes a phonon moving at the (average) speed of sound c_s to traverse thickness d, η is the probability that it will cross the interface, and the factor 4 comes from an angular average. Using $\eta = 0.25$ [22], $\tau_B(T/T_c = 0.78) \approx 8$ psec [10], and $\tau_s^{ph} \approx 80$ psec [10], we estimate $\tau_{es} \approx 180$ psec, $\tau_\gamma = 55$ psec, the phonon trapping enhancement factor $(1 + \tau_\gamma/\tau_B) = 8$, and we deduce $\tau_r = 22$ psec at $T/T_c = 0.78$. Kaplan [10] has calculated the temperature- and energy-dependent values of τ_r for Nb from an Eliashberg formulation and finds, for quasiparticles near the gap edge at $T/T_c = 0.78$, $\tau_r \approx 30$ psec. Because of uncertainties in the above parameters (for example, η is uncertain by $\pm 50\%$), the agreement between experiment and theory is quite good. Calculation also predicts that τ_r should have only a slight temperature dependence in the region $0.75 < T/T_c < 1.0$. Our samples systematically show a slight increase in τ_{eff} as T/T_c approaches 1.0, very similar to other reports [1]. At $T/T_c > 0.9$, $\delta N_{qp} \sim N_{qp} \geq N_p$ and the simplification expressed by Eq. (1) is not valid; τ_{eff} may be lengthened by enhanced pair breaking from critical-current and thermal-fluctuation effects.

The value of τ_r deduced from τ_{eff} is in good agreement with earlier results [8] of a branch-imbalance measurement of pure, bulk Nb. Based on the charge-imbalance lifetime τ_Q^0 measured in that experiment [9], a value $\tau_r = 60$ psec at $T/T_c = 0.75$ is expected. Furthermore, in

that experiment a biexponential decay was observed in a single-crystal sample. The ratio of long to short times, near T_c, was about 30. The present experiment has also observed a biexponential decay with a ratio τ_2/τ_{eff} of about 10. Although there is uncertainty in estimates of heat flow in the sample, substrate, and mounting configuration, τ_2 does not appear to be associated with any of these thermal time constants. For example, the small signals detected at $T > T_c$ should reveal any dominant thermal time constant. These signals decayed with characteristic times 0.8 ± 0.2 nsec that varied slightly with substrate but were independent of temperature. By contrast, τ_2 increases at $T \rightarrow T_c$ in approximately the same proportion as τ_{eff}, as did the signals of Ref. [8], and $\tau_2/\tau_{\text{eff}} \sim 10$ for all samples. Furthermore, τ_2 (and τ_{eff}) increased with increasing fluence in a manner consistent with a T^* model, but inconsistent with simple pictures of thermal decay. This suggests that biexponential decay is intrinsic to Nb.

In summary, we have applied a greatly improved technique for directly measuring quasiparticle lifetimes. The deduced recombination time is in very good agreement with theory. The observed biexponential decay in Nb films is in good agreement with previous measurement; however, the slow component remains unexplained within the present understanding of quasiparticle dynamics.

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