Effect of a magnetic field on the quasiparticle recombination in superconductors

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Quasiparticle recombination in a superconductor with an s-wave gap is typically dominated by a phonon bottleneck effect. We have studied how a magnetic field changes this recombination process in metallic thin-film superconductors, finding that the quasiparticle recombination process is significantly slowed as the field increases. While we observe this for all field orientations, we focus here on the results for a field applied parallel to the thin film surface, minimizing the influence of vortices. The magnetic field disrupts the time-reversal symmetry of the pairs, giving them a finite lifetime and decreasing the energy gap. The field could also polarize the quasiparticle spins, producing different populations of spin-up and spin-down quasiparticles. Both processes favor slower recombination; in our materials we conclude that strong spin-orbit scattering reduces the spin polarization, leaving the field-induced gap reduction as the dominant effect and accounting quantitatively for the observed recombination rate reduction.

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An excitation from the superconducting condensate requires finite energy (the energy gap $2\Delta$) and produces two quasiparticles. A quasiparticle excited to very high energy (compared to $\Delta$) quickly decays via a number of fast scattering processes to near the gap edge, where it recombines with a partner to form a Cooper pair. The pair’s binding energy is emitted mainly as $2\Delta$ phonons [1–3]. The recombination process is delayed by a phonon bottleneck: each recombination-generated phonon can break another Cooper pair, causing energy to be trapped in a coupled system of $2\Delta$ phonons and excess gap-edge quasiparticles [1–5]. Quasiparticle recombination has been widely studied in both metallic superconductors, to investigate the non-equilibrium processes in the many-body BCS system [7–10], and high-temperature superconductors, to gain new insight into the pairing mechanism [11–14]. Theories of the recombination process considered the reaction kinetics and interactions of quasiparticles and phonons [6, 16]; while experiments obtained the dependence of the quasiparticle lifetime on temperature, film thickness, and excitation strength [17, 18]. A magnetic field is known to couple to the electron orbital motion and to align the spin; both effects weaken superconductivity [19]. The consequence of magnetic field on the quasiparticle recombination [20] has not been examined in detail by optical pump-probe methods.

We use a novel time-resolved laser-pump synchrotron-probe spectroscopic technique to study the quasiparticle recombination dynamics in superconducting thin films, under applied magnetic field. Samples studied include a 10 nm thick Nb$_{0.5}$Ti$_{0.5}$N film on a crystal quartz substrate and a 70 nm thick NbN film on a MgO substrate. These substrates are essentially transparent in the far-infrared spectral range. The films were grown by reactive magnetron sputtering, using NbTi cathode in Ar/N$_2$ gas for Nb$_{0.5}$Ti$_{0.5}$N and Nb cathode in N$_2$ gas for NbN. The two films have critical temperatures of 10.2 K and 12.8 K, and a zero-temperature, zero-field, energy gap $2\Delta$ of 2.7 meV and 4.5 meV, respectively. Four-probe resistivity measurements with magnetic field parallel to the films determined

![FIG. 1. Experimental set-up. Electrons circulate in bunches in the synchrotron storage ring, generating pulses of far-infrared radiation with a repetition frequency of 52.9 MHz. The Ti:sapphire laser produces pulses with a repetition frequency of 105.8 MHz and a pulse picker selects every other pulse to match the synchrotron pulse pattern. The selected laser pulses are delivered over a fiber optic cable to the sample and the synchrotron pulse probes the photoinduced transmission at a fixed time delay afterward. To synchronize the synchrotron and laser pulses, the 52.9 MHz bunch timing signal from a pair of electrodes inside the synchrotron ring chamber is used by the Synchro-Lock laser control system as a reference for the laser pulse emission. The pulse generator introduces an adjustable delay between the laser and synchrotron pulses. The transmitted far-infrared light is detected by a bolometer detector and recorded on a computer.](image-url)
FIG. 2. Photoinduced transmission $S(t)$ vs. time $t$ for Nb$_{0.5}$Ti$_{0.5}$N (a and b) and for NbN (c and d), all measured in parallel fields at $T \lesssim 2$ K. Low-fluence and high-fluence data are compared. Note the semilog scale; simple exponential decay produces a straight line.

The samples were mounted in a $^4$He Oxford cryostat equipped with a 10 T superconducting magnet, and probed by far-infrared radiation produced in a bending magnet at beamline U4IR of the National Synchrotron Light Source, Brookhaven National Laboratory. The experiment, illustrated in FIG. 1, exploits the fact that the synchrotron radiation is emitted in $\sim$300 ps long pulses (governed by the electron bunch structure in the storage ring). We applied mode-locked near-infrared Ti:sapphire laser pulses ($\sim$2 ps in duration and $\sim$1.5 eV in photon energy) as the source for photoexcitation. The synchrotron probe beam measures the photoinduced optical properties due to the excess quasiparticles as a function of time delay relative to the arrival of the pump beam. The synchrotron pulse has a Gaussian profile with a FWHM of $\sim$300 ps, determining the time resolution of the experiment. At selected delay times $t$, we measure the spectrally integrated photoinduced transmission $S(t) \equiv -\Delta T(t)$ over the spectral range spanning the superconductors energy gap ($\sim$3 meV). The spectral shape is determined primarily by the optical components carrying the beam, and the detector.

If the laser were turned on and off to measure the photoinduced response, there would be a temperature modulation as well as the photoexcited quasiparticle modulation. To reduce these thermal effects we dither the laser pulse back and forth by a few tens of picoseconds at each delay setting, keeping the incident laser power constant. The dither is achieved by phase modulating the laser pulse using the internal oscillator of a lock-in amplifier. The directly obtained quantity is therefore a differential signal, $dS/ dt$. This signal was detected using a B-doped Si bolometer in combination with the lock-in amplifier. Numerical integration yields the photoinduced transmission $S(t)$, which directly follows the excess quasiparticle density $S$.

To study the effect of magnetic fields and excess carrier density on the recombination dynamics, we measured the magnetic-field and laser-fluence dependent photoinduced transmission for Nb$_{0.5}$Ti$_{0.5}$N and NbN thin films. The samples were fully immersed in superfluid $^4$He ($T \lesssim 2$ K) to minimize heating. At this low temperature, the thermal quasiparticle population is small (compared to the number of broken pairs at high fluence) but not zero. The field was applied parallel to the film surface to avoid the complexity of vortex effects. (See Ref. [3]). Typical results are shown in FIG. 2 where the photoinduced signal $S(t)$ (excess quasiparticle density) is plotted against delay time. At both low (FIG. 2a and FIG. 2c) and high laser fluences (FIG. 2b and FIG. 2d), a longer time is required for recombination as the magnetic field is increased. The pulse width of the synchrotron probe beam gives rise to the initial upturn in the data, which is skipped in the following data analysis.

We have discovered a revealing perspective to display our results, shown in FIG. 3. We define an effective instantaneous recombination rate $1/\tau_{\text{eff}}(t) \equiv -[dS(t)/ dt]/S(t)$ and plot $1/\tau_{\text{eff}}(t)$ vs $S(t)$ at various fields and fluences. Here short times are at the right (large $S(t)$) and long times at the left. In this presentation, data at the same field but for different pump fluences scale to the same straight line. As will be shown below, this behavior is expected for bimolecular recombination where the lifetime for a given particle is proportional to the availability of other particles with which to combine.

The scaling can be understood as follows. The phonon bottleneck was first discussed by Rothwarf and Taylor [1].
FIG. 4. Panels a and b show the excitation gap, $\Omega_G$ (squares) and the pair-correlation gap $\Delta$ (triangles) for Nb$_{0.5}$Ti$_{0.5}$N and NbN, obtained from the optical conductivity (left scale). The solid lines are theoretical calculations of $\Delta$ and $\Omega_G$. The square root of the condensate density $\sqrt{N_{sc}}$ (proportional to the order parameter) is shown as circles (right scale). Panels c and d show the slope extracted from Fig. 3 vs. $\Omega_G$ from a and b. The error bars in both plots are calculated deviations of the slope from the linear fit in Fig. 3. The lines are linear fits to the circles.

using two rate equations, one for the quasiparticles and the other for the $2\Delta$ phonons. The quasiparticles, which directly correspond to our signal $S(t)$, follow a simple model that captures the feature of bimolecular recombination, meanwhile taking into account the phonon bottleneck. The decay rate of the total quasiparticle density $N(t)$ toward the equilibrium density is proportional to $N^2$, because recombination requires the presence of two quasiparticles. Motivated by the Rothwarf-Taylor equations, we write

$$\frac{dN}{dt} = -2R(N^2 - N_{th}^2). \quad (1)$$

A thermal term $N_{th}^2$ is subtracted from $N^2$, because at equilibrium $N = N_{th}$ and the quasiparticle density must remain constant. The phonon bottleneck is introduced into the model through the recombination rate coefficient $R$. (See Section 1 of the Supplemental Material.) A factor of 2 is included because each recombination event depletes two quasiparticles. Now, $N(t) = N_{th} + N_{ex}(t)$, with $N_{th}$ the thermal density and $N_{ex}(t)$ the photoinduced excess density. At a given temperature and magnetic field, $N_{th}$ is time-independent, making Eq. (1) become

$$-\frac{dN_{ex}}{dt}/N_{ex} = 2R(N_{ex}(t) + 2N_{th}).$$

We identify $-\frac{dN_{ex}}{dt}/N_{ex}$ as the effective instantaneous relaxation rate $1/\tau_{eff}(t)$ defined earlier, because the photoinduced transmission $S(t)$ is proportional to the excess quasiparticle density $\sqrt{N_{ex}}$, where $C$ is just a constant to convert from signal to quasiparticle density. Hence,

$$-\frac{1}{S(t)} \frac{dS(t)}{dt} = \frac{2R}{C}(S(t) + 2S_{th}), \quad (2)$$

with $S_{th} = CN_{th}$. Eq. (2) is consistent with the linear behavior demonstrated in Fig. 3. The field dependence requires the prefactor $R$ to decrease with field.

To interpret the field dependence shown in Fig. 3 it is a prerequisite to understand how the field changes the electronic states of the superconductor. If spin-orbit scattering is small, the magnetic field could make the majority of quasiparticles have one spin direction. (This is the same polarization that gives Pauli paramagnetism to metals.) Spin polarization will slow the recombination because only quasiparticles with opposite spins can recombine. A recombination model including this spin polarization effect is discussed in Section 2 of the Supplemental Material. In this case, the recombination equation remains in the same form as Eq. (2), but with the coefficient $2R/C$ replaced by $(8R/C)(N_{↑}N_{↑}/N^2)$, where $N_{↑}$ and $N_{↓}$ are respectively the densities of spin-up and spin-down quasiparticles. The quasiparticle spin-polarization factor $N_{↑}N_{↓}$ would depend on the magnetic field in the limit of weak spin-orbit coupling, just as in the Pauli paramagnetism of metals. According to the BCS theory, electrons form singlet pairs condensed in the ground state; the spin susceptibility vanishes as the temperature approaches 0. The studies of superconductor spin susceptibility were done on thin films with thickness so small that the effect of a magnetic field on the electron orbit could be neglected. Paramagnetic splitting of the quasiparticle density of states was observed in 5 nm aluminum films in a parallel magnetic field [23]. In a study of magnetic field effects on far-infrared absorption of thin superconducting aluminum films, van Bentum and Wyder [24] concluded that paramagnetic splitting was important in their thinnest films, but did not allow for quasiparticle spin polarization. If a high degree of spin polarization existed, the recombination rate would be slowed.
much more than observed. However, spin-orbit scattering must be considered. Tedrow and Meservey observed the spin-state mixing in thin aluminum films due to spin-orbit scattering [25]. They defined a spin-orbit scattering parameter \( b = \hbar / 3 \Delta \tau_{so} \) to describe the degree of spin-orbit scattering, where \( \tau_{so} \) is the spin-orbit scattering time. They calculated that, as \( b \) is increased to 0.5, the spin-up and spin-down quasiparticle density of states completely mix, leaving no clear signature of the two-peak feature in the density of states due to Zeeman splitting. Considering the short spin-orbit scattering time measured [26] in NbTi, \( \tau_{so} = 3.0 \times 10^{-14} \text{s} \), and using the \( \Delta \) of Nb\(_{0.5}\)Ti\(_{0.5}\)N and NbN, we estimate that \( b = 4.2 \) and 3.3 for Nb\(_{0.5}\)Ti\(_{0.5}\)N and NbN, respectively. We believe that spin is not a good quantum number in our samples, requiring us to look beyond spin polarization to understand the recombination.

In a study [3] of the optical conductivity of Nb\(_{0.5}\)Ti\(_{0.5}\)N, we found that a parallel magnetic field breaks the time-reversal symmetry of the Cooper pairs and decreases the superconducting energy gap. The physics is similar to magnetic-impurity-induced pair-breaking effects, as originally formulated by Abrikosov and Gorkov [27]. In a magnetic field, one must distinguish between the spectroscopic energy gap \( 2\Omega_{G} \) and the pair-correlation gap \( \Delta \). These gaps [4] are plotted in FIG. 4a as squares and triangles respectively. The real part of the optical conductivity, corresponding to the electromagnetic absorption, shows a clear suppression of the energy gap \( 2\Omega_{G} \) with field (squares in FIG. 4a). The imaginary conductivity is a measure of the superconducting condensate density \( N_{sc} \), which goes as \( \Delta^2 \). The field dependences of \( \Delta \) and of \( \sqrt{N_{sc}} \) (shown as circles) agree well, providing clear evidence for a weakening of superconductivity by the magnetic field. The quantities \( \Omega_{G} \), \( \Delta \), and \( N_{sc} \) for NbN, obtained using the same technique (in Section 3 of the Supplemental Material), are plotted in FIG. 4b. The NbN field dependence is qualitatively different from that of Nb\(_{0.5}\)Ti\(_{0.5}\)N because in this thicker film the applied field induces a spatial variation in the order parameter, making the weakening of superconductivity proportional to the field, rather than being quadratic in field as in the much thinner Nb\(_{0.5}\)Ti\(_{0.5}\)N [29].

The energy gaps will be used in the following analysis.

The field dependence, shown in FIG. 3, is therefore field-dependent through \( \tau_{R} \), and is tied to the rates at which the phonons, produced in recombination events, re-break pairs (1/\( \tau_{B} \)) or escape from the film (1/\( \tau_{R} \)) [3]. The quasi-equilibrium values of \( \tau_{R} \) and \( \tau_{B} \) were derived by Kaplan et al. [6]. Magnetic-field-induced pair breaking decreases the spectroscopic energy gap (FIG. 4) and modifies the quasiparticle density of states, resulting in a decrease in \( \tau_{R} \) and an increase in \( \tau_{B} \). The field independent phonon escape time is determined by the film thickness and the acoustic mismatch between the film and the environment [32].

The recombination rate coefficient \( R \) (and, hence, the slope of 1/\( \tau_{eff} \) in FIG. 3) is therefore field-dependent through \( N_{th} \), \( \tau_{R} \) and \( \tau_{B} \). (See FIG. S7 in the Supplemental Material.) The equation is involved but, when we compute the slope vs \( \Omega_{G} \) for Nb\(_{0.5}\)Ti\(_{0.5}\)N and NbN, shown in FIG. 3a and FIG. 3b, we obtain a basically linear relation. This calculation implies a connection between the field-dependent quasiparticle recombination and the field-induced pair breaking. The linear relation can be explained by considering only the field-induced gap reduction. (See FIG. S7 in the Supplemental Material.) The finite y-intercepts in FIG. 4c and FIG. 4d are intriguing, bringing out the question of how the photoexcited quasiparticles relax in a gapless superconductor, motivating challenging experiments to probe the gapless regime.

In conclusion, our time-resolved pump-probe measurements on metallic \( s \)-wave superconductors reveal a slowing of quasiparticle recombination in an external magnetic field. The field was aligned parallel to the thin-film sample surface, to minimize effects due to vortices. There are two possible causes of the observed slowing: field-induced spin imbalance and field-induced gap reduction. The spin imbalance is unlikely to be important in Nb\(_{0.5}\)Ti\(_{0.5}\)N and NbN due to strong spin-orbit scattering. This scenario can be tested by investigating materials with small spin-orbit scattering. The field-induced gap reduction alone can explain quantitatively the slowing of recombination, and we conclude it to be the dominant effect observed in our experiment.

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Supplemental Material

1. Phonon bottleneck effect

In this section we show that the phonon bottleneck effect can be included in the effective recombination rate $R$ as a proportionality coefficient. Consider the Rothwarf-Taylor equations \cite{Rothwarf1970} for the coupled populations of quasiparticles and phonons in the absence of external quasiparticle injection,

$$
\frac{dN}{dt} = -\tau^{-1}R N^2 + \tau_B^{-1}N_w, \quad (S1)
$$

$$
\frac{dN_w}{dt} = \frac{1}{2} \tau^{-1}R N^2 - \frac{1}{2} \tau_B^{-1}N_w - \tau^{-1}_\gamma (N_w - N_w,\text{th}), \quad (S2)
$$

where $N(t)$ and $N_w(t)$ are respectively the densities of quasiparticles and high-energy (defined as $h\omega \geq 2\Delta$) phonons, $\tau^{-1}_R$ is the intrinsic quasiparticle recombination rate, $\tau_B^{-1}$ is the phonon pair-breaking rate, and $\tau^{-1}_\gamma$ is the phonon escape rate. (The phonons may enter the substrate, enter the helium bath, or decay anharmonically to energies $h\omega < 2\Delta$.) In thermal equilibrium, the quasiparticles $N(t)$ and high-energy phonons $N_w(t)$ reach time-independent equilibrium values, linked by $N_{\text{th}} = \sqrt{N_w,\text{th}\tau_B^{-1}/\tau^{-1}_R}$, where $N_{\text{th}}$ and $N_w,\text{th}$ are for the quasi-particles and high-energy phonons, respectively. The coupled non-linear equations (S1) and (S2) can be solved numerically, e.g., using the Runge-Kutta integration method. To illustrate the solutions, we set $\tau^{-1}_R = 1$ ns$^{-1}$ and $\tau_B^{-1} = 10$ ns$^{-1}$, typical values for these quantities at low temperatures \cite{Rothwarf1969}. Without loss of generality we set $N_w,\text{th} = 1$, which determines $N_{\text{th}} = \sqrt{10}$. The initial value of $N$ is determined by the pump laser fluence. We consider three cases, $N(0) = 2N_{\text{th}}$ for low fluence, $N(0) = 5N_{\text{th}}$ for intermediate fluence, and $N(0) = 10N_{\text{th}}$ for high fluence. $N_w(0)$ is set to $N_w,\text{th}$. For each case, we consider a range of phonon escape rates ($\tau^{-1}_\gamma$) from 1 ns$^{-1}$ to 10 ns$^{-1}$, spanning the strong to the weak phonon-bottleneck regime.

The numerical solutions of the Rothwarf-Taylor equations are shown in the first row of FIG. S1. After a short period of approximately 0.05 ns, the phonon bottleneck effect becomes clear. As expected, when the phonon escape rate increases, the phonon bottleneck effect becomes weaker and recombination becomes faster. In the second row of FIG. S1 the rate of change of the excess quasiparticle density $N_{\text{ex}}(t) = N(t) - N_{\text{th}}$ is plotted vs. $N_{\text{ex}}$. The early stage ($t < 0.05$ ns) shown in the first row of FIG. S1 has been skipped, because the quasiparticle and phonon populations are not yet equilibrated and it does not give information about the phonon bottleneck. Moreover, this stage is not temporally resolved in our experiment.

The quasi-linear relation shown in the second row of FIG. S1 suggests we can define an effective recombination rate $R$ as $-(dN_{\text{ex}}/dt)/N_{\text{ex}}$. This rate, defined as the slope of $-dN_{\text{ex}}/dt$ vs. $N_{\text{ex}}$, is plotted vs. $\tau^{-1}_\gamma$ in FIG. S2. As $N(0)$ increases from $2N_{\text{th}}$ to $10N_{\text{th}}$, the relation between $R$ and $\tau^{-1}_\gamma$ becomes almost linear. We estimated that, for our experimental conditions, $N(0)$ in our samples was three orders of magnitude greater than $N_{\text{th}}$ at the lowest laser fluence. Therefore it is safe to conclude that the effective recombination rate $R$ scales linearly with the phonon escape rate $\tau^{-1}_\gamma$.

2. Pauli paramagnetism

Our recombination model can be extended to include the magnetic-field dependence due to quasiparticle spin polarization. In a quasiparticle recombination event, both a spin-up and a spin-down quasiparticle are needed to form a Cooper pair. We use an equation similar to the band-to-band recombination equation in a semiconductor to describe the quasiparticle recombination,

$$
\frac{dN}{dt} = \frac{dN^\uparrow}{dt} + \frac{dN^\downarrow}{dt} = -8R(N^\uparrow N^\downarrow - N_{\text{th}}^\uparrow N_{\text{th}}^\downarrow), \quad (S3)
$$

where $\uparrow$ and $\downarrow$ denote the spin-up and spin-down populations, respectively. This reduces to Eq. (1) in the main text when the spin-up and spin-down populations are equal. At a given condition, Eq. (S3) can be rewritten as $-(dN_{\text{ex}}/dt)/N_{\text{ex}} = 8R(N^\uparrow N^\downarrow)(N_{\text{ex}} + 2N_{\text{th}})/N^2$, where $N_{\text{ex}} = N - N_{\text{th}}$. Relating the measured photocarried transmission $S$ to the excess quasiparticle density, $S = CN_{\text{ex}}$, where $C$ is a constant, the recombination equation can be further reduced to

$$
-\frac{1}{S} \frac{dS}{dt} = 8R \frac{N^\uparrow}{CN^2}(N^\uparrow N^\downarrow)(S + 2S_{\text{th}}), \quad (S4)
$$

where $S_{\text{th}} = CN_{\text{th}}$.

The majority spin fraction $N^\uparrow/N$ can be calculated from the paramagnetic model, assuming Fermi-Dirac distribution of quasiparticles $f(E)$ and the quasiparticle density of states $D(E)$ from the BCS theory,

$$
N^\uparrow = 2 \int_0^\infty f(E)D(E + \mu H) dE, \quad (S5)
$$

$$
N^\downarrow = 2 \int_0^\infty f(E)D(E - \mu H) dE. \quad (S6)
$$

If we assume that the field dependence we see in FIG. 3 in the main text is only through the product $N^\uparrow N^\downarrow$, we can extract the majority spin fraction at different fields. The results are compared with the calculation in FIG. S3. Pure Pauli paramagnetism predicts a stronger magnetic field dependence than observed in our data.

The theory, however, must consider the strong spin-orbit scattering in NbN and Nb$_{0.5}$Ti$_{0.5}$N. In the main text, we estimate the strong spin-state mixing in the Nb$_{0.5}$Ti$_{0.5}$N and Nb$_{0.5}$Ti$_{0.5}$N samples. Based on that argument and the analysis shown in this section, we expect that the spin polarization factor $N^\uparrow N^\downarrow$ is weakly dependent on the field for our samples.
3. Analysis of optical conductivity for NbN

We studied the magnetic-field-induced effects in the Nb$_{0.5}$Ti$_{0.5}$N and NbN thin films using Fourier transform far-infrared spectroscopy. The experimental technique and the analysis for Nb$_{0.5}$Ti$_{0.5}$N can be found in Ref. [3]. Here we analyze the superconducting-state to normal-state transmission ratio $T_s/T_n$ and reflection ratio $R_s/R_n$ for NbN shown in FIG. S4, measured at 2 K with the magnetic field parallel to the film. The data were taken with 4 cm$^{-1}$ (0.5 meV) resolution, so that the fringes due to the multiple internal reflections in the substrate were not resolved. The angle of incidence for both transmission and reflection was 30°. The NbN thin film has a normal-state
conductivity $\sigma_n = 2.0 \times 10^3 \text{ Ohm}^{-1}\text{cm}^{-1}$, determined from its normal-state transmittance and thickness. The MgO substrate has a refractive index $n \approx 3.0$ and negligible absorption in the far-infrared. The zero-field gap $\Delta_0 = 17.9 \text{ cm}^{-1}$ is obtained from fitting the zero-field optical conductivity with the Mattis-Bardeen theory.

From the transmission and reflection ratios, we extracted the real ($\sigma_1$) and imaginary ($\sigma_2$) parts of the optical conductivity using the method discussed in Ref. [3]. The results are shown in FIG. S5. We found that the field dependence can be explained well by the pair-breaking theory. (See details of the theory in Ref. [4].)

The pair-breaking parameter $\Gamma$ is the only fitting parameter, describing the strength of pair breaking due to the magnetic field. Its value at different fields is shown in FIG. S6. From $\Gamma$ we calculated the pair-correlation gap $\Delta$ and the effective spectroscopic gap $G$ using $\ln(\Delta/\Delta_0) = -\pi\Gamma/4\Delta$ and $G = \Delta[1 - (\Gamma/\Delta)^{2/3}]^{1/2}$. These quantities are shown in FIG. 4b in the main text. The superconducting condensate density $N_{sc}$ is estimated from the below-gap part of $\sigma_2$ at $T \ll T_c$, which has the form,

$$\sigma_2(\omega) = \frac{N_{sc}e^2}{m \omega}, \quad (S7)$$

where $e$ and $m$ are the electron charge and mass, respectively.

4. Exact solution of the recombination equation

The recombination equation, Eq. (2) in the main text, links the measured $S(t)$ to the model,

$$-\frac{1}{S(t)} \frac{dS(t)}{dt} = \frac{2R}{C}(S(t) + 2S_{th}),$$

where $S_{th} = CN_{th}$. This equation has the following exact solution:

$$S(t) = S(0) \frac{2e^{-t/\tau}}{2 + S(0)/S_{th}(1 - e^{-t/\tau})}. \quad (S8)$$

Here $\tau$ is the effective lifetime; $\tau = 1/4RN_{th}$. In the low-fluence regime, especially when $S(0) \ll S_{th}$, the solution is close to an exponential decay. As the fluence increases, the deviation from a simple exponential decay becomes significant.

5. Effective recombination rate

On the one hand, the quasiparticles, interacting with phonons in the system, decay with an effective lifetime $\tau_{eff} \approx \tau_{\gamma} + (1/2)\tau_R(1 + \tau_\gamma/\tau_B)$, where $\tau_{\gamma}$, $\tau_R$, and $\tau_B$ are the same quantities as defined above and in the main
At low temperatures, $\tau_R \gg \tau_\gamma$ and $\tau_\gamma \gg \tau_B$. The effective lifetime can be approximated as $\tau_{\text{eff}} = \tau_R \tau_\gamma / 2\tau_B$. On the other hand, by solving the recombination equations proposed in the main text, one can identify an effective quasiparticle lifetime $\tau_{\text{eff}} = 1/4RN_{th}$, as shown in the previous section. As a result,

$$R = \frac{\tau_B}{2\tau_\gamma \tau_R N_{th}} = \frac{\tau_\gamma^{-1} \tau_R^{-1}}{2\tau_B^{-1} N_{th}}.$$  \hfill (S9)

The phonon escape rate $\tau_\gamma^{-1}$ is expected to be independent of the gap, as discussed in the main text. A theory for $\tau_R^{-1}$ and $\tau_B^{-1}$ has been given by Kaplan et al. \cite{Kaplan}.

$$\tau_R^{-1}(\omega) = \frac{\tau_0^{-1}}{(k_BT_e)^3} \int_0^\infty \frac{\Omega^2(\Omega - \omega)}{\sqrt{\omega^2 - \Delta^2}} [n(\Omega) + 1] f(\Omega - \omega) d\Omega, \hfill (S10)$$

$$\tau_B^{-1}(\Omega) = \frac{\tau_{0,\text{ph}}^{-1}}{\pi \Delta(0)} \int_\Delta^{\pi/2} \frac{\Omega - \omega}{\sqrt{\omega^2 - \Delta^2}} d\omega, \hfill (S11)$$

where $\tau_0^{-1}$ and $\tau_{0,\text{ph}}^{-1}$ are respectively the characteristic lifetimes of the quasiparticles and phonons, determined by the electron-phonon coupling and phonon density of states. The quantities $f$ and $n$ are the Fermi-Dirac and Bose-Einstein distribution functions, respectively. $\tau_R^{-1}$ should be evaluated at the quasiparticle gap energy $\omega = \Delta$ and $\tau_B^{-1}$ should be evaluated at the phonon energy $\Omega = 2\Delta$ for the estimation of their near-equilibrium values. For $\tau_R^{-1}$

$$\tau_R^{-1}(\Delta) \approx \frac{\tau_0^{-1}}{(k_BT_e)^3} \int_{2\Delta}^{\infty} d\Omega \frac{\Omega^5 e^{-(\Omega - \Delta)/k_BT}}{\Delta} = \frac{\tau_0^{-1} e^{-\Delta/k_BT}}{(k_BT_e)^3} \int_0^\infty dx \frac{(x + 2\Delta)^{5/2}}{x^{3/2} e^{-x/k_BT}}, \hfill (S12)$$

in which we have replaced the Bose factor $n(\Omega)$ and the Fermi factor $f(\Delta)$ by their low-temperature values. Because only small $x$ in the integrand contributes significantly to the integral, an expansion of the numerator yields

$$\tau_R^{-1}(\Delta) \approx \frac{\tau_0^{-1} e^{-\Delta/k_BT}}{(k_BT_e)^3} (2\Delta)^{5/2} \left[ \int_0^\infty dxx^{-1/2} e^{-x/k_BT} + \frac{5}{4\Delta} \int_0^\infty dxx^{3/2} e^{-x/k_BT} \right] = \frac{\tau_0^{-1} e^{-\Delta/k_BT}}{(k_BT_e)^3} (2\Delta)^{5/2} 2\pi k_BT \left( 1 + \frac{5k_BT}{8\Delta} \right). \hfill (S13)$$

The phonon pair-breaking rate $\tau_B^{-1}$ has a simple form for its near-equilibrium state, given in Ref. \cite{Kaplan} as

$$\tau_B^{-1}(2\Delta) = \tau_{0,\text{ph}}^{-1} \frac{\Delta}{\Delta_0} (1 - 2f(\Delta)) \approx \tau_{0,\text{ph}}^{-1} \frac{\Delta}{\Delta_0}. \hfill (S14)$$

The quasiparticle density is

$$N_{th} \approx N(0) \sqrt{2\pi \Delta_0 k_BT} e^{-\Delta/k_BT}. \hfill (S15)$$

Substituting Eqs. (S13)–(S15) into Eq. (S9) yields

$$R \approx \frac{2\sqrt{2\Delta_0}}{N(0)} \frac{\tau_\gamma^{-1} \tau_\tau^{-1}}{\tau_{0,\text{ph}}^{-1} (k_BT_e)^3} \left( 1 + \frac{5k_BT}{8\Delta} \right). \hfill (S16)$$

In the context of pair breaking, $\Delta$ should everywhere be replaced by the field-dependent spectroscopic gap $\Omega_G$.

The gap dependence of $\tau_R$, $\tau_B$, $N_{th}$, and $R$ can also be numerically evaluated directly using Eqs. (S10), (S11), (S15), and (S9) without approximations. The results are shown in Fig. S7, confirming the linear relation between $R$ and $\Omega_G$, discussed in the main text and shown in Fig. 4c and 4d.

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