Recombination-Limited Energy Relaxation in a Bardeen-Cooper-Schrieffer Superconductor

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We study quasiparticle energy relaxation at subkelvin temperatures by injecting hot electrons into an Al island and measuring the energy flux from quasiparticles into phonons both in the superconducting and in the normal state. The data show strong reduction of the flux at low temperatures in the superconducting state, in qualitative agreement with the theory for clean superconductors. However, quantitatively the energy flux exceeds the theoretical predictions both in the superconducting and in the normal state, suggesting an enhanced or additional relaxation process.

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Superconducting nanostructures attract lots of attention, partly because of their potential applications, for instance, in single Cooper pair and single-electron devices, in quantum information processing, and in detection of radiation. Although the operation of many of these devices is based on charge transport, the energy relaxation is also of importance to warrant proper functioning either under driven conditions or when subjected to environment fluctuations. Thermalization of the quasiparticle system with the surrounding bath is a serious concern at subkelvin temperatures for nonsuperconducting structures, but securing proper thermalization of a superconductor is an even greater challenge. Recombination of hot quasiparticles (qp's) into Cooper pairs slows down exponentially towards low temperatures. Quasiparticle scattering rates in usual superconductors have been assessed theoretically several decades ago [1,2] and measured experimentally both soon after the first predictions [1] and also recently at very low temperatures [3–5]. However, the associated heat flux in superconductors has not been addressed in the past. This is the topic of the present Letter. We present both experimental and theoretical results which demonstrate the importance of slow thermal relaxation in superconducting nanostructures.

Energy relaxation in normal metals has been investigated thoroughly for a long time [6–9]. In three-dimensional systems, quasiparticle-phonon (qp-ph) heat flux $P_{\text{qp-ph}}$ is

$$P_{\text{qp-ph}} = \Sigma \mathcal{V}(T_{\text{qp}}^5 - T_{\text{ph}}^5). \tag{1}$$

Here Σ is a material constant [10], $\mathcal{V}$ is the volume of the system, and T_{qp} and T_{ph} are the temperatures of quasiparticles and phonons, respectively. Deviations from this behavior towards the fourth power of temperature have been seen for lower temperatures [9,11] and are usually explained by the impurity effects [12–14] when the wavelength of a thermal phonon becomes longer than the quasiparticle mean free path or of the sample size. Nevertheless, Eq. (1) gives a good account of the heat flux for most experiments at subkelvin temperatures. Under the same conditions, quasiparticle-quasiparticle (qp-qp) relaxation is typically much faster; most experiments demonstrate the so-called quasiequilibrium, where qp's have a well-defined temperature, usually different from that of the phonons. Deviations from this picture have been observed, e.g., in voltage biased diffusive wires [15].

Relaxation processes in superconductors have also been studied [1]. The most obvious distinctions from the normal state are (i) the qp's need to emit or absorb an energy in excess of the gap Δ to be recombined or excited, and (ii) the number of qp's is very small well below the critical temperature (T_C). This all leads to exponentially slow qp-ph relaxation rates at low temperatures. The relaxation rate was addressed recently in experiments on superconducting detectors [3–5]; these measurements suggest to confirm the recombination-limited rate $\tau_{\text{rec}}^{-1} \propto \sqrt{T/T_C} e^{-\Delta/k_B T}$ down to $T/T_C \simeq 0.2$. At lower T, the relaxation rate saturates due to presently poorly known reasons.

For clean superconductors, the qp-ph energy flux can be derived in the spirit of Eq. (1) using the electron-phonon matrix elements from the quasiclassical theory [16]:

$$\begin{aligned} P_{\text{qp-ph}} = &-\frac{\Sigma \mathcal{V}}{96\zeta(5)k_B^5} \int_{-\infty}^{\infty} dE E \\ &\times \int_{-\infty}^{\infty} d\epsilon \epsilon^2 \text{sgn}(\epsilon) L_{E,E+\epsilon}\left[\coth\left(\frac{\epsilon}{2k_B T_{\text{ph}}}\right)\right. \\ &\left.\times (f_E^{(1)} - f_{E+\epsilon}^{(1)}) - f_E^{(1)} f_{E+\epsilon}^{(1)} + 1\right]. \end{aligned} \tag{2}$$

Here $f_E^{(1)} = f(-E) - f(E)$; $f(E)$ is the distribution function of qp's; it is the Fermi function $f_{T_{\text{qp}}}(E) = (1 + e^{E/k_B T_{\text{qp}}})^{-1}$ if qp's are in equilibrium at temperature T_{qp}. Phonons are assumed to be in equilibrium with occupation $n_{\text{ph}}(\mathbf{q}, T_{\text{ph}}) = (e^{\epsilon_{\mathbf{q}}/k_B T_{\text{ph}}} - 1)^{-1}$ at temperature T_{ph}. The factor $L_{E,E'} = N_{T_{\text{qp}}}(E) N_{T_{\text{qp}}}(E')[1 - \frac{\Delta^2(T_{\text{qp}})}{EE'}]$. Here $N_{T_{\text{qp}}}(E) = |E|/\sqrt{E^2 - \Delta(T_{\text{qp}})^2} \Theta(E^2 - \Delta(T_{\text{qp}})^2)$ is the superconducting density of states (DOS) normalized to the normal-metal DOS at the Fermi level $\nu(E_F)$ [$\Theta(x)$ is the Heaviside step function]. We obtain $P_{\text{qp-ph}} \simeq \frac{64}{63\zeta(5)} \Sigma \mathcal{V} T_{\text{qp}}^5 e^{-\Delta/k_B T_{\text{qp}}}$ for $T_{\text{ph}} \ll T_{\text{qp}} \ll \Delta/k_B$, which is by a factor $0.98 e^{-\Delta/k_B T_{\text{qp}}}$ smaller than in the normal state [with $T_{\text{ph}} \ll T_{\text{qp}}$].

In the present measurements, we aim at realizing the situation discussed above when the electrons injected into the island are in quasiequilibrium at a temperature T_{qp} decoupled from the heat bath which consists of thermal phonons at much lower temperature T_{ph}. The power absorbed in the island is then associated with the heat transferred to phonons emitted by thermal qp's. Figure 1 shows a typical configuration of our experiments. The samples were made by electron beam lithography and shadow evaporation of aluminum in two angles. Reported results have been obtained at the conditions when the junctions are either superconducting (S, I, S, I for insulator) or normal (N, I, N). Hybrid junctions between S and N were not used, since they are not sensitive in probing S. The parameters of the structures are given in Table I. The aluminum block in the center of Fig. 1 is the volume in which energy relaxation is investigated. Two small and two large tunnel junctions connect the island to aluminum leads (thickness 40 nm). The hot qp's are injected via one of the small tunnel junctions in series with a large one. Because of the large asymmetry of junction parameters, essentially all of the power is injected by the small junction. The steady-state distribution on the island is deduced from the current-voltage (I-V) curves of the opposite pair of junctions. We observe the qp current of only the small junction; the large junction remains in the supercurrent state. Measurements in a configuration with two small junctions in series as injectors and the two large junctions in series as probes were also made with essentially identical results.

If the island is at temperature T_{qp} and the lead is at T_{ext}, the qp current I in opaque tunnel junctions is given by $eR_T I = \int dE N_{T_{\text{qp}}}(E - eV) N_{T_{\text{ext}}}(E)[f_{T_{\text{qp}}}(E - eV) - f_{T_{\text{ext}}}(E)]$, where R_T is the junction resistance. For $T_{\text{qp}} = T_{\text{ext}}$, (a) calculated and (b) measured I-V curves for various T_{qp}/T_C are shown in Fig. 2. Wide plateaus in the regime $0 < eV < 2\Delta$ emerge due to the thermal qp current; its

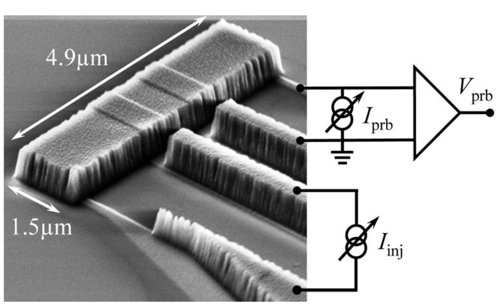

FIG. 1. A typical sample (sample C) for measuring energy relaxation in an Al superconducting bar. The circuits indicate injection of hot qp's and probing the island temperature.

value at $eV = \Delta$ is shown in Fig. 2(c). The agreement between experiment and theory is good down to $T_{\text{qp}}/T_C \simeq 0.25$. Therefore, and since the estimated power input due to the probing current is orders of magnitude smaller than that due to injection, the temperature increase due to measurement is assumed to be vanishingly small. To match the data to the theory also at lower temperatures one can use the pair-breaking parameter $\gamma \equiv \Gamma/\Delta$ resulting in a smeared DOS: $N_{T_{\text{qp}}}(E) = |\text{Re}(E + i\Gamma)/\sqrt{(E + i\Gamma)^2 - \Delta(T_{\text{qp}})^2}|$. In the figure we show lines with $\gamma = 10^{-4}$ and $\gamma = 10^{-3}$. We focus our analysis to the range $0.3 < T_{\text{qp}}/T_C < 1$ where no fit parameter is needed.

At $T \sim T_C$ the qp-qp and the qp-ph relaxation rates for aluminum films are [10,17,18] $\gamma_{\text{qp-qp}} \sim 10^8$–$10^9$ s^{-1} and $\gamma_{\text{qp-ph}} \sim 10^6$–$10^7$ s^{-1}, respectively. Even for energies of the order of injection voltage $eV \sim 100 k_B T_C$, we have $\gamma_{\text{qp-qp}} > \gamma_{\text{qp-ph}}$ [19] which ensures nearly thermal qp distribution in our samples. The deviation from quasiequilibrium produced by injection through a tunnel contact is, on one hand, determined by the effective rate $\eta = 1/4\nu(E_F)e^2 \mathcal{V} R_T$ [20]. In our samples (see Table I) the contacts with high tunnel resistance $R_T \sim 1$ MΩ have $\eta \sim 10$ s^{-1}. For low-resistance contacts which have a much lower voltage drop, $\eta \sim 10^3$ s^{-1}. These values are much smaller than both $\gamma_{\text{qp-qp}}$ and $\gamma_{\text{qp-ph}}$. Therefore this condition of quasiequilibrium is well satisfied. To warrant quasiequilibrium the scattering rate of qp's should also be not smaller than the recombination rate. Based on the data of Ref. [5] and the theory [1], the two rates can become comparable in our experiment. However, the favorable comparison in Fig. 2 between the calculated thermal I-V curves and those measured under power injection shows that our samples are nearly in quasiequilibrium. Moreover, there may exist effects of nonequilibrium phonons, as well [21]. Yet such phonons would lead to a deviation between theory and experiment, which is of opposite sign to what we will present (Fig. 3).

Figure 2(d) shows the calculated I-V curves of the probe junction, assuming that only the island temperature T_{qp} is elevated and the leads remain at $T_{\text{ext}} = 0.05 T_C$. This is the expected behavior in quasiequilibrium under power injection, provided the junctions are opaque enough not to conduct heat from the island into the leads. A peak in the I-V curves arises at $eV = \Delta(T_{\text{ext}}) - \Delta(T_{\text{qp}})$. In Fig. 2(e), we show the corresponding measured curves at various levels of injected power. The resemblance between Figs. 2(d) and 2(e) is obvious and supports the adopted picture of thermal distribution of injected qp's. Note that

TABLE I. Sample dimensions and junction resistances.

Sample	Volume (μm^3)	R_1, R_2, R_3, R_4 (kΩ)
A	$21 \times 1.5 \times 0.44$	840, 4, 4, 1160
B	$4.9 \times 1.5 \times 0.44$	760, 5.7, 5.7, 1290
C	$4.9 \times 1.5 \times 0.44$	485, 20, 20, 980

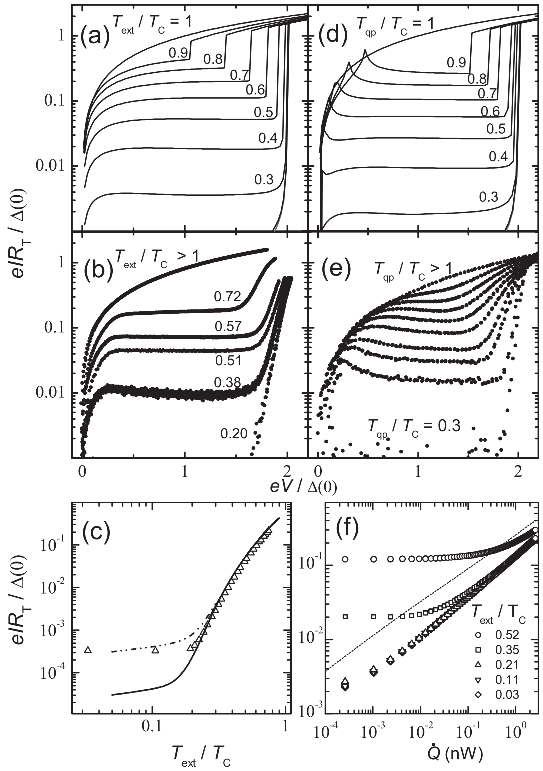

FIG. 2. Tunnel currents for a superconductor in equilibrium and quasiequilibrium. (a) Theoretical and (b) experimental I-V curves of a junction at several bath temperatures when $T_{qp} = T_{ext}$ (sample A). (c) Theoretical and experimental currents at $eV = \Delta$ (sample B). The two theory lines correspond to pair-breaking parameters $\gamma = 10^{-3}$ (upper curve) and $\gamma = 10^{-4}$ (lower curve). (d) Calculated I-V curves when the leads and the island have different temperatures $T_{qp} \neq T_{ext}$. (e) The measured I-V curves under a few injection conditions (sample C). (f) The current in sample A on the plateau between the initial peak and the rise of the current at the conduction threshold around $2\Delta/e$. The theoretical prediction for the lowest T_{ext} is shown by the dashed line. The value of Δ at zero temperature is 200 ± 5 μeV, and $T_C = 1.45 \pm 0.03$ K.

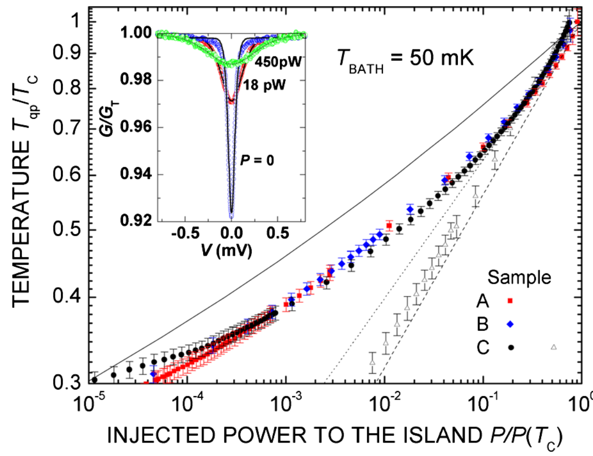

FIG. 3 (color online). Energy relaxation from theory and experiment. The data in the superconducting state are from samples A (squares), B (diamonds), and C (circles). The open triangles are from sample C in the normal state. The solid line is the result of Eq. (2) in the superconducting state. The dotted line indicates $P/P(T_C) = (T_{qp}/T_C)^5$, and the dashed line $P/P(T_C) = (T_{qp}/T_C)^4$. The inset shows three Coulomb peaks measured in the normal state under different levels of power injection; the solid lines are theoretical fits to them.

the features in experimental curves are broadened which is common for small junctions (see [22] and references therein) but can also originate from a finite qp lifetime (nonzero Γ) in the superconductors. In the data analysis we next find the minimum current in the plateaulike regime at bias voltages between the ''matching'' peak and the strong onset of qp current. This current is converted into temperature by comparing it to the T_{qp}-dependent minimum current of the theoretical I-V curves.

The expression for power deposited on the island by a biased junction $\dot{Q}(V) = (e^2R_T)^{-1}\int(E - eV)N_{T_{qp}}(E - eV)N_{T_{ext}}(E)[f_{T_{ext}}(E) - f_{T_{qp}}(E - eV)]dE$ allows us to determine the injected power and the heat flux through all of the junctions. We note two features: (i) Since injection voltages in the experiment are $V \gg \Delta/e$, it is sufficient to assume that the power injected into the island equals $IV/2$, i.e., it is divided evenly between the two sides of the junction. (ii) The heat flux through the (probing) junction is given by the equation in the beginning of this paragraph with unequal T_{ext} and T_{qp}. It is almost constant over a wide range of voltages within the gap region. Its value is low and can be neglected under most experimental conditions. Yet, to test this, we varied the resistances of the large tunnel junctions by a factor of 5 between samples A and C, without a significant effect on the results. Figure 2(f) shows the current on the plateau as a function of power injected, at various bath temperatures. In a wide range, from 30 up to 380 mK, the behavior is almost identical: The power depends only on the higher temperature between T_{qp} and T_{ph}, consistent with the theoretical discussion. Therefore we compare the experimental results at the base phonon temperature (of about 50 mK) to the theory predictions for $T_{ph} \ll T_{qp}$ in what follows.

We studied $P_{qp\text{-}ph}$ in the normal state as well by applying a magnetic field of about 120 mT to suppress the superconductivity and measuring the partial Coulomb blockade (CB) signal [23]. Like in the superconducting state, two regimes are possible. In equilibrium the results of Ref. [23] apply. Under injection, the typical situation is such that $T_{ext} \ll T_{qp}$, which we discuss now in more detail. The tunneling rates in a state with an extra charge n for adding (+) or removing (−) a qp to or from the normal island with electrostatic energy change $\Delta F^{\pm}(n) = \pm 2E_C(n \pm 1/2) \mp eV/2$ are

$$\Gamma^{\pm}(n) = \frac{1}{e^2R_T}\int_{-\infty}^{\infty} dEf_1(E)\{1 - f_2(E - \Delta F^{\pm}(n))\}. \quad (3)$$

Here $E_C = e^2/2C_\Sigma$ is the charging energy of the island with the total capacitance C_Σ, and f_1 and f_2 are the distributions on the source and target electrodes. For equilibrium distribution $f_i(E) = (1 + e^{E/k_B T_i})^{-1}$, with $T_1 = T_2$, Eq. (3) yields the result of Ref. [23]. Here we have the opposite limit of low bath temperature $T_{\rm ext} = T_1 \ll T_2 = T_{\rm qp}$. For $T_1 = 0$, $f_1(E) = 1 - \Theta(E)$, yielding $\Gamma^{\pm}(n) = (k_B T_{\rm qp}/e^2 R_T) \ln(1 + e^{-\Delta F^{\pm}(n)/k_B T_{\rm qp}})$. The current into the island is $I = e\sum_{n=-\infty}^{\infty} \sigma(n)[\Gamma^+(n) - \Gamma^-(n)]$, where $\sigma(n)$ is the probability of having n extra qp's on the island. Since $\sum_{n=-\infty}^{\infty} n\sigma(n) = 0$ by symmetry, and $\sum_{n=-\infty}^{\infty} \sigma(n) = 1$, we find for the differential conductance up to the first order in $E_C/k_B T_{\rm qp}$

$$\frac{G^{\rm neq}}{G_T} = 1 - \frac{E_C}{2k_B T_{\rm qp}} \frac{1}{\cosh^2(eV/4k_B T_{\rm qp})}. \tag{4}$$

The depth of the conductance minimum at $V = 0$ is $\Delta G/G_T = E_C/2k_B T_{\rm qp}$, which is 50% larger than that in the equal-temperature case. The full width at half minimum is $V_{1/2}^{\rm neq} = 4\ln(3 + 2\sqrt{2})k_B T_{\rm qp}/e$. This is about 65% of the equal-temperature value $V_{1/2}^{\rm eq} \simeq 10.88 k_B T_{\rm qp}/e$ [23].

Figure 3 is a collection of the data at the base temperature ($\simeq$50 mK), in the form of island temperature $T_{\rm qp}/T_C$ as a function of injected power. The superconducting state was measured for the three samples. The power is normalized by that at T_C, to present data from different samples on the same footing. For samples A, B, and C, $P(T_C) = 14$, 3, and 3 nW, respectively. The data on the three samples are mutually consistent. The superconductor result Eq. (2) is shown by a solid line. The normal state data were taken for sample C, which is ideal for a measurement of the island temperature via partial CB: It has $E_C/k_B \simeq 20$ mK (see the inset in Fig. 3). The two large junctions were used for probing and the small ones for power injection. We first checked that the value $V_{1/2}^{\rm eq}$ yields a good quantitative agreement with the equilibrium temperature data over the whole range of the experiment. Next, we measured the qp temperature under injection. The low base temperature permits the use of the expression of $V_{1/2}^{\rm neq}$ above to extract $T_{\rm qp}$ in the range displayed in Fig. 3. Power-law-type behavior can be observed over the whole temperature range $0.3T_C < T_{\rm qp} \lesssim T_C$. The data approach those of the superconducting state near $T_C \simeq 1.45$ K, as expected. The power law for $P_{\text{qp-ph}}$ is, however, better approximated by $T_{\rm qp}^4$ (dashed line) instead of $T_{\rm qp}^5$ (dotted line) of Eq. (3), yielding a deviation of the same sign with respect to the basic theories as in the superconducting state.

The data demonstrate that qp-ph coupling in a superconductor is weaker than in the normal state, by 2 orders of magnitude at $T_{\rm qp}/T_C = 0.3$. But, like in the relaxation time experiments in a superconductor [3–5], the energy flux is larger than that from the theory [1,16]. This observation could suggest that the qp relaxation rate both in the superconducting and in the normal state might be sensitive to the microscopic quality and the impurity content of the particular film [14]. The impurity effects on the qp-ph relaxation are controlled by the parameter $q\ell$, where ℓ is the qp mean free path and $q = k_B T_{\rm qp}/\hbar u$ is the wave vector of an emitted phonon with energy of the order of the qp temperature. With the speed of sound $u \sim 5000$ m/s and $\ell \sim 20$ nm in our samples, we have $q\ell \sim 0.5{\rm K}^{-1} T_{\rm qp}$. Thus, the impurity effects can become essential below 1 K.

Our experiments on three samples with very different parameters yielded essentially identical results when normalized by the island volume. Thus, we believe that issues such as thermal gradients, nonequilibrium, charge imbalance, and heat leaks through tunnel contacts have only a minor influence on the results. The data thus yield the intrinsic energy relaxation of qp's in the superconducting and in the normal state. In summary, the experiment follows qualitatively the theoretical model that we presented. Quantitatively, there is a substantial discrepancy especially for superconductors, which would imply that one needs to invoke an extra relaxation channel to account for.

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