

**Quasiparticle number fluctuations in superconductors**

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We present a general theory of quasiparticle number fluctuations in superconductors. The theory uses the master equation formalism. First, we develop the theory for a single occupation variable. Although this simple system is insufficient to describe fluctuations in a physical superconductor, it is illustrative, allowing this discussion to serve as a self-contained introduction. We go on to develop a multivariate theory that allows for an arbitrary number of levels with transitions of arbitrary size between levels. We specialize the multivariate theory for two particular cases. First, we consider intrinsic quasiparticle fluctuations. In a previous Letter, these results were used to describe time-resolved measurements of thermodynamic fluctuations in a superconducting Al box [C.M. Wilson, L. Frunzio, and D.E. Prober, *Phys. Rev. Lett.* **87**, 067004 (2001)]. Finally, we extend these results to include fluctuations due to extrinsic loss processes.

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**I. INTRODUCTION**

Superconductivity is a rich, physical phenomenon with many aspects that have been studied for their possible technological importance. The most basic property of superconductors, their ability to transport electrical currents without resistance, has been applied broadly for many years. A new generation of superconducting electronic devices aims to take advantage of more subtle aspects of superconductivity, including flux quantization, quantum tunneling and the quantum coherence of the superconducting state. Examples include superconducting quantum interference devices, high-speed electronics,<sup>1</sup> superconducting detectors,<sup>2</sup> and various implementations of quantum bits for quantum information processing.<sup>3</sup> The ultimate sensitivity and usefulness of these devices will be determined in part by the physical processes that add noise to them.

In this article, we present a theory of one such noise source: fluctuations in the number of quasiparticle excitations. In its ground state, all of the conduction electrons in a superconductor form bound pairs, called Cooper pairs. The binding energy of the pairs is the spectroscopic gap  $E_g = 2\Delta$ , where  $\Delta$  is the energy gap for a single excitation. At finite temperature, some pairs will be broken, resulting in single-particle excitations known as quasiparticles. In equilibrium, the average number of quasiparticles  $N^0$  is determined by thermodynamics. In particular, the average occupation of quasiparticle levels is determined by the Fermi-Dirac distribution, with the energy measured from the Fermi energy  $\varepsilon_F$  and the minimum quasiparticle energy being  $\Delta$ . At a microscopic level, it is the balance of quasiparticle generation and recombination that determines the average number of quasiparticles. Quasiparticle generation refers to the creation of two quasiparticle excitations when a Cooper pair is broken by a thermal phonon. Quasiparticle recombination refers to the annihilation of two quasiparticles as they form a Cooper pair and emit a phonon. Generation and recombination are random processes, meaning that individual generation or recombination events occur at random intervals. Because of this, the instantaneous density of quasiparticles fluctuates in time. Statistical mechanics tells us that the r.m.s.

magnitude of the fluctuations is  $(N^0)^{1/2}$  (Ref. 4). In a recent Letter, we confirmed the prediction for the magnitude and we demonstrated that, at low temperatures, the time scale of the fluctuations is the recombination time  $\tau_R^*$ .<sup>5</sup>

In this paper, we present a general theory of quasiparticle fluctuations in superconductors. (Previous treatments of quasiparticle fluctuations were restricted to basic thermodynamic arguments.<sup>6</sup>) We use the master equation formalism, which has been used to describe fluctuations in semiconductors for many years. The master equation formalism reproduces and expands the thermodynamic results, while also being applicable to nonequilibrium systems. In Sec. II, we develop the theory for a single occupation variable. This simple system is insufficient to describe fluctuations in a physical superconductor, but we have included it because it is illustrative, allowing the article to serve as a self-contained introduction to researchers in superconductivity that are unfamiliar with the semiconductor research. In addition, the more complete theory will show that the simpler results of the one-variable system can be used with the appropriate definition of effective parameters. In Sec. III, we develop a multivariate theory that allows for an arbitrary number of levels with transitions of arbitrary size between levels. We then specialize the multivariate theory to two particular cases. In Sec. III B, we consider intrinsic quasiparticle fluctuations where quasiparticles are only created (annihilated) in pairs due to thermal generation (recombination). In a previous Letter, this specialized case was used to describe time-resolved measurements of thermodynamic fluctuations in a superconducting Al box.<sup>5</sup> In Sec. III C, we also allow quasiparticles to be lost and created individually. This second case can be applied to systems with normal metal traps, diffusive loss, etc.

**II. SINGLE-VARIABLE MASTER EQUATION**

To treat fluctuations in our system, we construct a master equation similar to the Fokker-Planck equation. This differential equation describes the probability distribution of the occupancies of various subsystems (levels). We follow the treatment by van Vliet of generation-recombination noise in

semiconductors,<sup>7</sup> except that we generalize the description to allow for transitions that involve an arbitrary number of particles, e.g., two quasiparticles recombining. The master equation formalism can in fact predict the fluctuations of an arbitrary number of coupled levels. However, that development is not particularly illuminating. For this reason, we will start with the derivation for a two-level system described by a one-variable master equation.

We can consider one level of our system to be quasiparticles. The second level could be Cooper pairs or quasiparticles in traps or something else, depending on the exact nature of the system that we are trying to model. In this section, we will refer to any processes that creates (annihilates) quasiparticles as a generation (recombination) process, although in general these terms have the specific meanings defined in Sec. I. Regardless of exactly what the second level is, it is not in general independent of the first level because the total number of excitations in the two levels is constrained. For instance, the number of quasiparticles plus Cooper pairs is constrained by the total number of electrons, due to overall charge neutrality. Furthermore the creation of two quasiparticles implies the loss of one pair, and vice versa. Therefore, we only need to count the number of quasiparticles  $N$  and can describe our system with a one variable master equation

$$\begin{aligned} \frac{\partial P(N, t|k, 0)}{\partial t} = & -[g(N) + r(N)]P(N, t|k, 0) + g(N) \\ & - \delta N P(N - \delta N, t|k, 0) + r(N + \delta N)P(N \\ & + \delta N, t|k, 0), \end{aligned} \quad (1)$$

where  $P(N, t|k, 0)$  is the probability that there are  $N$  quasiparticles at time  $t$  given that there were  $k$  quasiparticles at  $t=0$ . The function  $g(N)$  is the probability per unit time that there will be a generation event in the box when there are  $N$  quasiparticles. In other words,  $g(N)dt$  is the probability of a generation event in the time interval  $dt$ . Similarly, the function  $r(N)$  describes the probability per unit time of recombination. The parameter  $\delta N$  is the number of quasiparticles added (removed) by a generation (recombination) event. We can understand the structure of the master equation quite simply. It describes the rate of change of the probability that there are  $N$  quasiparticles in the system. The rate of decrease in the probability equals the probability that there are  $N$  quasiparticles times the probability per unit time that there will be a generation or recombination event. This is what the first term in the master equation represents. The rate of increase in the probability is equal to the probability that the system is one generation event away from having  $N$  quasiparticles times the probability per unit time that there will be a generation event, plus a similar term for recombination.

The master equation is a countably infinite set of coupled differential equations. Luckily, we do not need to solve the master equation for it to be useful. We can instead use the master equation to construct much simpler equations for quantities such as the variance and correlation function of the fluctuations.

We begin by calculating the variance of the fluctuations. The variance is a steady-state property, so we can set the left

side of the master equation to zero. If we then multiply the equation by  $N$ , and sum over all possible  $N$ , we get the simple relationship

$$\langle g(N) \rangle = \langle r(N) \rangle,$$

where the angle brackets mean the expectation value over all  $N$ . If we expand both  $g(N)$  and  $r(N)$  in a Taylor expansion in  $N$  around the equilibrium value  $N^0$ , we get

$$g(N^0) + \frac{1}{2}g''(N^0)\langle \Delta N^2 \rangle = r(N^0) + \frac{1}{2}r''(N^0)\langle \Delta N^2 \rangle, \quad (2)$$

where the primes indicate the derivative with respect to  $N$  and  $\Delta N = N - N^0$ . The first order terms vanish because  $\langle \Delta N \rangle = 0$  in equilibrium. In addition, in most cases  $g(N), r(N) \propto N^2$ , and  $\langle \Delta N^2 \rangle \propto N$ , so we can neglect the second order terms and simplify to

$$g(N^0) \approx r(N^0).$$

This is the reasonable statement that the generation and recombination rates must balance in equilibrium.

If we again set the left-hand side of the master equation (1) to zero, multiply by  $N^2$  and sum over all  $N$ , we get the relationship

$$\left\langle \left( N + \frac{\delta N}{2} \right) g(N) \right\rangle = \left\langle \left( N - \frac{\delta N}{2} \right) r(N) \right\rangle.$$

If we again expand  $g(N)$  and  $r(N)$  around  $N^0$  and use Eq. (2) to simplify, we can find the following expression for the variance of the fluctuations:

$$\langle \Delta N^2 \rangle = \delta N \frac{r(N^0)}{r'(N^0) - g'(N^0)}, \quad (3)$$

where we have again neglected second order terms in the final expression.

We can also use the master equation to calculate the power spectrum of the fluctuations. To do this, we first calculate the autocorrelation function of the fluctuations and then compute its Fourier transform. The autocorrelation function at lag  $u$  is defined as

$$\Phi(u) = \langle N(0)N(u) \rangle = \sum_k \sum_j k_j P(k, 0; j, u),$$

where  $P(k, 0; j, u)$  is the *joint* probability that there are  $k$  quasiparticles at  $t=0$  and that there are  $j$  quasiparticles at  $t=u$ . (By lag we mean the amount of time that one signal is shifted with respect to the other.) We can simplify this expression by factoring the joint probability distribution into  $P(k, 0; j, u) = P(j, u|k, 0)P(k, 0)$  giving

$$\Phi(u) = \sum_k k P(k, 0) \sum_j j P(j, u|k, 0) = \sum_k k \langle N \rangle_k P(k, 0), \quad (4)$$

where  $P(j, u|k, 0)$  is the conditional probability of having  $j$  quasiparticles at  $t = u$  given that there were  $k$  at  $t = 0$  and  $\langle N \rangle_k$  is the expectation value of  $N$  given that there were  $k$  quasiparticles at  $t = 0$ .

To further simplify this expression, we start by deriving a differential equation for  $\langle N \rangle_k$  using the master equation. In this case, we need to use the full master equation (1) without setting the time derivative equal to zero. If we multiply both sides by  $N$  and sum over all  $N$ , we get the equation

$$\frac{d}{du} \langle N \rangle_k = \delta N (\langle g(N) \rangle - \langle r(N) \rangle).$$

We cannot solve this equation explicitly, because we do not know the expectation values on the right-hand side. However, we can find an approximate solution by again expanding  $g(N)$  and  $r(N)$  around  $N^0$ . We find the simple result

$$\begin{aligned} \frac{d}{du} \langle \Delta N \rangle_{k-N^0} &= - \frac{\langle \Delta N \rangle_{k-N^0}}{\tau}; \\ \tau &\equiv \frac{1}{\delta N} \frac{1}{r'(N^0) - g'(N^0)}, \end{aligned} \quad (5)$$

where  $\tau$  appears as the effective relaxation time of the fluctuations. This equation has the simple solution

$$\langle \Delta N \rangle_{k-N^0} = (k - N^0) \exp\left(-\frac{u}{\tau}\right).$$

Inserting this solution into Eq. (4) we find the autocorrelation function of the fluctuations to be

$$\Delta \Phi(u) = \langle \Delta N(0) \Delta N(u) \rangle = \langle \Delta N^2 \rangle \exp\left(-\frac{u}{\tau}\right),$$

where  $\langle \Delta N^2 \rangle$  is the variance of the fluctuations. We can then directly compute the power spectrum  $G(\omega)$  of the fluctuations as the Fourier transform of the autocorrelation function. We find

$$G(\omega) = \frac{4 \langle \Delta N^2 \rangle \tau}{1 + \omega^2 \tau^2}.$$

We now have general expressions for the variance and power spectrum of the fluctuations in our two-level system. Before we specialize the equations more, we can make some general comments. First, if we combine Eq. (3) with Eq. (5), we find the much simpler expression for the variance of the fluctuations

$$\langle \Delta N^2 \rangle = (\delta N)^2 r(N^0) \tau.$$

This says that the variance of  $N$  is of order the number of particles that recombine in one correlation time. Now, looking at Eq. (5), we see that  $\tau$  is inversely proportional to  $\delta N$ . This says the more quasiparticles that are lost (created) by a single recombination (generation) event, the faster the fluctuations. Also, looking at Eq. (5) we see that the time scale of fluctuations is inversely proportional the sum of the derivatives of the generation and recombination rates. This has a

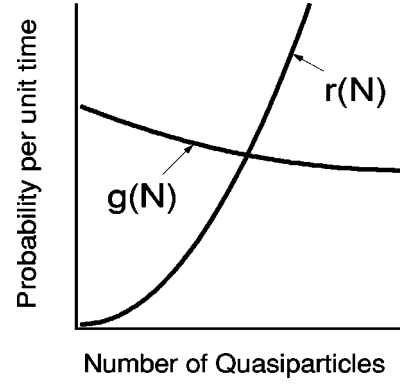


FIG. 1. Sketch of the generation and recombination parameters  $g(N)$  and  $r(N)$ . The intersection of the curves yields the steady-state number of quasiparticles.

simple physical interpretation. In Fig. 1 we sketch the recombination parameter  $r(N)$  and generation parameter  $g(N)$  as a function of  $N$ . First, we note that the value of  $N$  where the curves intersect is the equilibrium value  $N^0$ . Next, we notice that for a stable system the derivative of  $r(N)$  will always be positive and the derivative of  $g(N)$  will always be negative. This is what maintains equilibrium. For example, if  $N$  fluctuates greater than  $N^0$ , then the recombination rate increases *and* the generation rate decreases. Both of these changes drive the system back to equilibrium. Even more, the steeper the change in the rates around equilibrium, the faster the system is driven back to equilibrium. This is why the time constants depend on the derivatives of  $r(N)$  and  $g(N)$  and why their contributions sum together.

To be able to apply the formulas derived above we must know what  $r(N)$  and  $g(N)$  are for our system. Luckily, if we already understand the dynamics of the system, it is general easy to deduce  $r(N)$  and  $g(N)$ . In general, the rate equation of our system will be of the form

$$\frac{dN}{dt} = \delta N [g(N) - r(N)]. \quad (6)$$

If we can derive or know an appropriate rate equation for our system, we can then read off  $g(N)$  and  $r(N)$ .

We can consider, as an example, the case of simple generation and recombination of quasiparticles. By simple, we mean that quasiparticles are only lost to recombination with other quasiparticles and we ignore the effects of phonon trapping (which we will return to later). In this case, the two levels of our system are quasiparticles and Cooper pairs, with the total number of electrons constrained to be the normal state value. We will further assume that we are working at low temperatures and that the number of quasiparticles is small compared to the number of Cooper pairs. In general, we would expect  $g(N)$  to depend on the number of Cooper pairs. However, since the relative size of the fluctuations will be small compared to the number of Cooper pairs, we will assume  $g(N)$  is constant and equal to the equilibrium recombination rate. With that we can write the rate equation for our simple system as

$$\frac{dN}{dt} = 2 \left( \Gamma_G - \frac{1}{2} \frac{R}{\text{vol}} N^2 \right),$$

where  $\Gamma_G$  is the constant generation rate, vol is the volume of the system, and  $R$  is the recombination constant. The recombination constant is basically a constant of proportionality between the recombination rate and the number of ways to combine  $N$  quasiparticles, which is  $N^2/2$ .

From this rate equation, we can read off the parameters of our model

$$g(N) = \Gamma_G, \quad r(N) = \frac{1}{2} \frac{R}{\text{vol}} N^2, \quad \delta n = 2.$$

We can then easily put these parameters into the equation above to find a familiar result for the variance of the fluctuations  $\langle \Delta N^2 \rangle = N^0$ . We can also easily write down the power spectrum of the fluctuations

$$G(\omega) = \frac{4N^0\tau}{1 + \omega^2\tau^2}, \quad \tau = \frac{\text{vol}}{2RN^0}. \quad (7)$$

We see that the spectrum has a simple Lorentzian form with a bandwidth given by  $1/\tau$ .

### III. MULTIVARIABLE MASTER EQUATION

#### A. General theory

The simple one-variable master equation derived above is illustrative, but it is not sufficient to describe generation and recombination in a physical superconductor. For example, in

$$Q(\mathbf{a}; \mathbf{a}'') = \begin{cases} p_{ij}, & \mathbf{a}'' = \{N_1, \dots, N_i, \dots, N_j, \dots\} \\ & \mathbf{a} = \{N_1, \dots, N_i - \delta n_{ij}, \dots, N_j + \delta n_{ji}, \dots\} \\ 0, & \text{otherwise} \end{cases},$$

where  $\delta n_{ij}$  is the ‘‘shot size.’’ The physical meaning of  $\delta n_{ij}$  is the change in the occupation of level  $i$  when making a transition to or from level  $j$ . This is one important generalization of the master equation formalism for superconductors. In typical semiconductor systems, transitions between all levels change the occupation by one, i.e.,  $\delta n_{ij} = \delta n = 1$  for all transitions. In superconductors, however, not only can different levels have a different shot size, they can have a different shot size depending on what the other level involved in the transition is.

We can then proceed along the same lines as the derivation in Sec. II. We will not include the detailed derivation, instead presenting the results and referring to Ref. 7 for a more detailed treatment. In analogy to the linearized time constant found in Eq. (5), we can write a linearized rate matrix  $M$ , where the elements are

$$M_{ij} = \sum_k \delta n_{ik} \left( \frac{\partial p_{ik}}{\partial N_j} - \frac{\partial p_{ki}}{\partial N_j} \right) \Bigg|_{\{N_i\} = \{N_i^0\}}. \quad (9)$$

a thin-film superconductor the phonon emitted when a pair of quasiparticles recombines can break another pair before the phonon escapes the film into the bath. This process, known as phonon trapping, extends the effective lifetime of a quasiparticle. To account for this process, or others similar to it, we must increase the number of levels in our model system. The basic idea is the same as before, except we now describe the state of the system with levels (1)–( $S$ ) by a vector  $\mathbf{a} = (N_1, N_2, \dots, N_S)$  which represents the occupation of each level. In general only  $S - 1$  levels will be independent since the total number of excitations is constrained. We start by writing down the master equation for the system:

$$\frac{\partial P(\mathbf{a}, t | \mathbf{a}', 0)}{\partial t} = \sum_{\mathbf{a}'' \neq \mathbf{a}} P(\mathbf{a}'', t | \mathbf{a}', 0) Q(\mathbf{a}; \mathbf{a}'') - \sum_{\mathbf{a}'' \neq \mathbf{a}} P(\mathbf{a}, t | \mathbf{a}', 0) Q(\mathbf{a}'', \mathbf{a}), \quad (8)$$

where  $P(\mathbf{a}, t | \mathbf{a}', 0)$  is the probability that the system is in state  $\mathbf{a}$  at time  $t$  given that it was in state  $\mathbf{a}'$  at  $t=0$ , etc., and  $Q(\mathbf{a}; \mathbf{a}'')$  is the transition probability per unit time from state  $\mathbf{a}''$  to  $\mathbf{a}$ . Again, the first term says that the rate of change in the probability of finding the system in state  $\mathbf{a}$  is the probability of it being one transition away from  $\mathbf{a}$  times the rate of transition to  $\mathbf{a}$ . The second term accounts for transitions out of state  $\mathbf{a}$ . We can make this less abstract if we notice that the only allowed transitions in our system involve a single loss event in one level causing a creation event in a second level. We can then write

We can define a second matrix  $\mathbf{B}$  (which describes the second order Fokker-Plank moments) whose elements are

$$B_{ii} = \sum_{k \neq i} \delta n_{ik}^2 (p_{ki} + p_{ik}) \approx 2 \sum_{k \neq i} \delta n_{ik}^2 p_{ik}^0,$$

$$B_{ij} = -\delta n_{ij} \delta n_{ji} (p_{ij} + p_{ji}) = -\delta n_{ij} \delta n_{ji} (p_{ij}^0 + p_{ji}^0). \quad (10)$$

The covariance matrix  $\sigma^2 = \langle \Delta \mathbf{a} \cdot \Delta \mathbf{a}^T \rangle$ , is then determined by the following matrix equation:

$$\sigma^2 \cdot \mathbf{M}^T + \mathbf{M} \cdot \sigma^2 = \mathbf{B}, \quad (11a)$$

where  $\Delta \mathbf{a} = \mathbf{a} - \mathbf{a}^0$ . We can also write the cross power spectrum matrix as

$$\mathbf{G}(\omega) = 2 \text{Re}[(\mathbf{M} + i\omega \mathbf{1})^{-1} \mathbf{B} (\mathbf{M}^T - i\omega \mathbf{1})^{-1}], \quad (12a)$$

where  $\text{Re}[\dots]$  means the real part and  $\mathbf{1}$  is the identity matrix. The diagonal terms of  $\mathbf{G}$  describe the power spectrum of the



fluctuations of each level in the system. The off-diagonal terms of  $\mathbf{G}$  describe the cross power spectrum between the various levels. Each spectrum  $G_{ij}$  is a sum of individual Lorentzian spectra, similar to Eq. (7), with characteristic frequencies determined by the eigenvalues of  $\mathbf{M}$ .

At this point, we wish to add some general comments. First, we note that in the Fokker-Plank approximation to the master equation,  $\mathbf{M}$  would describe the ‘‘drift’’ of the system and  $\mathbf{B}$  would describe the ‘‘diffusion.’’ In this context, Eq. (11a) can be viewed as a generalized Einstein relation, connecting drift (mobility) and diffusion.<sup>8</sup> Second, we note that these results implicitly assume that our system is linear with respect to fluctuations, meaning that the fluctuations do not drive our inherently nonlinear system out of the regime of linear response. (This property is also referred to as quasilinearity.) If this assumption is violated, then a different formalism must be used.<sup>8</sup>

Equations (11a)-(12a) can be simplified for some systems, including equilibrium systems. Specifically, they can be simplified in systems that have a symmetric correlation matrix, i.e., systems, where

$$\langle \Delta \mathbf{a}(t) \cdot \Delta \mathbf{a}^T(0) \rangle = \langle \Delta \mathbf{a}(0) \cdot \Delta \mathbf{a}^T(t) \rangle.$$

If this condition holds, then we can demonstrate that  $\sigma^2 \cdot \mathbf{M}^T = \mathbf{M} \cdot \sigma^2$ , and Eqs. (11a) and (12a) reduce to

$$\sigma^2 = \langle \Delta \mathbf{a} \cdot \Delta \mathbf{a}^T \rangle = \frac{1}{2} \mathbf{M}^{-1} \cdot \mathbf{B} \quad (11b)$$

and

$$\mathbf{G}(\omega) = \frac{2}{\omega^2} \text{Re} \left[ \left( \mathbf{1} + \frac{\mathbf{M}}{i\omega} \right)^{-1} \mathbf{B} \right]. \quad (12b)$$

Note that the symmetry of the correlation matrix requires that both the correlators  $\langle \Delta N_i(t) \Delta N_i(0) \rangle$  and the cross-correlators  $\langle \Delta N_i(t) \Delta N_j(0) \rangle$  exhibit time-reversal symmetry, i.e.,  $\langle \Delta N_i(t) \Delta N_j(0) \rangle = \langle \Delta N_i(0) \Delta N_j(t) \rangle$ . For classical particles (meaning that the  $\{n_i\}$  are numbers and not operators), the symmetry of the correlators is trivial. However, the cross correlators are not required to be symmetric. In an equilibrium system, the cross correlators *are* symmetric as a result of the time-reversal symmetry of the underlying microscopic dynamics (microscopic reversibility).<sup>8</sup> Therefore, the simplified results (11b) and (12b) can always be used in equilibrium. They also approximately apply to systems in quasiequilibrium, which we define as a steady-state condition that obeys the principle of detailed balance, i.e.,  $p_{ij}^0 = p_{ji}^0$  for all  $i$  and  $j$  (Ref. 9). In equilibrium, detailed balance is a consequence of microscopic reversibility, but it does not generally apply to systems in nonequilibrium steady state. In steady state, we have the more general relationship  $\sum_{i \neq j} p_{ij}^0 = \sum_{i \neq j} p_{ji}^0$ , which simply means that the total transition rate into a level must balance the total transition rate out. However, depending on the details of the level structure, this more general relationship can reduce to the expression for detailed balance even in steady state. In particular, this reduction can apply in steady-state systems where levels are coupled in pairs (see Sec. III C).

An important special case of Eq. (12b) is the quasiequilibrium, two-variable result. Since we are always free to label the quasiparticles as level 1, we will give the general expression for  $G_{11}$  in the two variable case

$$G_{11}(\omega) = 2 \sum_{1,2} \frac{\tau_1 \tau_2}{(\tau_2 - \tau_1)} \frac{\tau_1^2}{[1 + (\omega \tau_1)^2]} \left[ \left( \frac{1}{\tau_1} - M_{22} \right) B_{11} + M_{12} B_{12} \right], \quad (13)$$

where  $\gamma_i = 1/\tau_i$  are the eigenvalues of  $\mathbf{M}$  and the summation means add another term with  $\tau_1$  and  $\tau_2$  interchanged. The result for  $G_{22}$  has the same form but with the indices 1 and 2 interchanged on the components of  $\mathbf{M}$  and  $\mathbf{B}$ .

We will not discuss in detail any examples of systems driven out of quasiequilibrium, but a thorough discussion of such systems can be found in Ref. 9. Reference 9 discusses a three level system driven through a cycle of transitions: (1) photoexcitation of carriers, (2) trapping of carriers, and (3) recombination in the traps. It is found that this driven system can exhibit super-Poissonian noise, i.e.,  $\langle \Delta N^2 \rangle \gg N^0$ , while quasiequilibrium systems can only exhibit sub-Poissonian noise, i.e.,  $\langle \Delta N^2 \rangle \leq N^0$ .

## B. Intrinsic quasiparticle fluctuations

The first specific example that we will consider is intrinsic quasiparticle fluctuations in a thin-film superconductor. By intrinsic fluctuations we mean (1) that quasiparticles are only created in pairs through generation, whereby a Cooper pair is broken by a high-energy phonon and (2) quasiparticles are only lost in pairs through recombination, whereby a Cooper pair is formed with the emission of a high-energy phonon. This system can be described by three levels whose populations are labeled by  $N$ ,  $N_\omega$ , and  $N_{\omega,B}$  which are the number of quasiparticles in the superconducting electrode, the number of phonons with energy  $E_\omega > 2\Delta$  in the superconducting electrode, and the number of phonons with  $E_\omega > 2\Delta$  in the bath respectively. We only keep track of phonons with  $E_\omega > 2\Delta$  because they are the only phonons that can generate new quasiparticles. In thin-film systems, the ‘‘bath’’ would generally be the substrate on which the superconducting electrode is fabricated. The important distinction between phonons in the electrode and phonons in the bath is that phonons in the bath cannot generate quasiparticles.

In Sec. II, we thought of two quasiparticles recombining to form a Cooper pair, instead of quasiparticles recombining to form a phonon. In the end, however,  $N_\omega$  is a more natural variable than the number of Cooper pairs for several reasons. From a statistical point of view, we can account for the recombination of two quasiparticles equally well as a transition to a Cooper pair or a transition to a phonon. From a dynamical point of view, however, keeping track of phonons is much more important than keeping track of Cooper pairs. As we will see shortly, the presence of phonons created by recombination can significantly change the effective recombination rate measured in experiments. On the other hand, the rate  $\Gamma_B$  at which phonons break pairs and generate quasipar-

icles  $is$  proportional to the number of Cooper pairs, but as long as the number of pairs is much greater than the number of quasiparticles, then  $\Gamma_B$  is approximately constant. Thus, we see that  $N_\omega$  is a better choice.

We can describe the dynamics of the levels with the following system of three coupled differential equations:

$$\frac{dN}{dt} = 2 \left\{ -\frac{1}{2} \frac{RN^2}{\text{vol}} + \Gamma_B N_\omega \right\}, \quad (14)$$

$$\frac{dN_\omega}{dt} = \frac{1}{2} \frac{RN^2}{\text{vol}} - \Gamma_B N_\omega - \Gamma_{\text{es}} N_\omega + \Gamma_K N_{\omega,B}, \quad (15a)$$

$$\frac{dN_{\omega,B}}{dt} = \Gamma_{\text{es}} N_\omega - \Gamma_K N_{\omega,B}, \quad (16)$$

where  $\Gamma_{\text{es}}$  is the rate at which phonons escape from the electrode to the bath and  $\Gamma_K$  is the rate at which phonons enter the electrode. We have neglected the anharmonic decay of phonons as a loss process because it happens on a time scale much longer than phonon escape at these energies.

We can simplify these equations with the approximation that  $N_{\omega,B}$  is constant, which is justified because the exchange of phonons with the electrode is a very small perturbation to the bath. This simplification reduces Eq. (16) to the equality  $\Gamma_{\text{es}} N_\omega^0 = \Gamma_K N_{\omega,B}^0$ , where the superscripts indicate steady-state values. We can then rewrite Eq. (15a) as

$$\frac{dN_\omega}{dt} = \frac{1}{2} \frac{RN^2}{\text{vol}} - \Gamma_B N_\omega - \Gamma_{\text{es}} (N_\omega - N_\omega^0). \quad (15b)$$

We then see that Eqs. (14) and (15b) are the well known Rothwarf-Taylor equations.<sup>10</sup>

Following Gray,<sup>11</sup> we can linearize these equations for small perturbations by writing  $N = N^0 + \Delta N$  and  $N_\omega = N_\omega^0 + \Delta N_\omega$  and simplifying. If we define the vector  $\mathbf{a} = (N, N_\omega)$  then we can write the linearized equations in matrix form

$$\frac{d(\Delta \mathbf{a})}{dt} = -\Gamma \cdot \Delta \mathbf{a}, \quad \Gamma = \begin{pmatrix} 2\Gamma_R & -2\Gamma_B \\ -\Gamma_R & \Gamma_\omega \end{pmatrix}, \quad (17)$$

where we have taken  $\Gamma_\omega = \Gamma_B + \Gamma_{\text{es}}$  and  $\Gamma_R = RN^0/\text{vol}$  as the steady-state recombination rate. The eigenvalues of  $\Gamma$  determine the time constants of the system's response to small perturbations. Gray showed that the dominant time constant for the quasiparticle response in the limit  $\Gamma_R \ll \Gamma_B + \Gamma_{\text{es}}$  is

$$\Gamma_R^* = 2\Gamma_R F_\omega^{-1}, \quad F_\omega = 1 + \frac{\Gamma_B}{\Gamma_{\text{es}}}, \quad (18)$$

where  $F_\omega$  is called the phonon trapping factor. It accounts for a phonon emitted by a recombination event breaking another pair before it escapes to the bath. We note that  $F_\omega^{-1}$  is just the probability that a phonon escapes to the bath.  $\Gamma_R^*$  is the time constant with which a small perturbation of the quasiparticle system will decay, and it is the rate we expect to measure in experiments. We see that the measured recombination rate  $\Gamma_R^*$  is generally very different from the true equilibrium recombination rate  $\Gamma_R$ . Even for  $F_\omega = 1$ , we see that  $\Gamma_R^*$

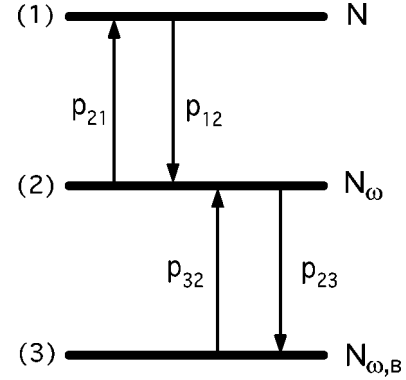


FIG. 2. Schematic representation of our three level system. From top to bottom, the levels are quasiparticles, phonons in the electrode, and phonons in the bath.

$= 2\Gamma_R$ . This is a consequence of quasiparticles recombining in pairs. [Formally, it arises from the linearization of the  $N^2$  term in Eq. (14).]

We can now specialize the multivariable master equation to describe the fluctuations in our intrinsic system. Our three levels are connected by various transitions labeled  $\{p_{ij}\}$  in Fig. 2. Each transition represents a physical process that changes the occupation of the three levels. Transition  $p_{12}$  describes two quasiparticles recombining to create one phonon in the electrode. Transition  $p_{21}$  describes the reverse process, a phonon being absorbed and generating two quasiparticles. Transition  $p_{23}$  describes a phonon escaping from the electrode into the bath. Finally,  $p_{32}$  describes a phonon entering the electrode from the bath. We note that there is no direct connection between levels 1 and 3, the quasiparticles and the bath. Since we have a three level system, our underlying master equation is a two variable equation. We choose as our two variables the number of quasiparticles  $N$  and the number of phonons in the electrode  $N_\omega$ . Referring to the rate equations for the system (14)–(16) we can read off the transition probabilities, which we tabulate in Table I.

In addition to the transition probabilities, we can also read off the shot size for each level, which is  $\delta n_1 = 2$  for the quasiparticles and  $\delta n_2 = 1$  for the phonons. Plugging these parameters into the above equations we find

$$\mathbf{M} = \begin{pmatrix} 2\Gamma_R & -2\Gamma_B \\ -\Gamma_R & \Gamma_\omega \end{pmatrix}, \quad \mathbf{B} = \Gamma_R N^0 \begin{pmatrix} 4 & -2 \\ -2 & 1 + \frac{\Gamma_{\text{es}}}{\Gamma_B} \end{pmatrix}, \quad (19)$$

TABLE I. Allowed transitions and the probability per unit time for each one.

Transition	Symbol	Probability per unit time
recombination	$p_{12}$	$(1/2)RN^2/\text{vol}$
generation	$p_{21}$	$\Gamma_B N_\omega$
phonon escape	$p_{23}$	$\Gamma_{\text{es}} N_\omega$
phonon entry	$p_{32}$	$\Gamma_{\text{es}} N_\omega^0$

where  $\Gamma_\omega = \Gamma_{\text{es}} + \Gamma_B$ . With these matrices we can then write the covariance matrix for our system. We find

$$\sigma^2 = \begin{pmatrix} N^0 & 0 \\ 0 & N_\omega^0 \end{pmatrix} = \begin{pmatrix} N^0 & 0 \\ 0 & \frac{1}{2} \frac{\Gamma_R}{\Gamma_B} N^0 \end{pmatrix},$$

where we have used the principle of detailed balance to relate  $N_\omega^0$  to  $N^0$ . Thus, we again find that the variance of the occupation of each level is equal to the average occupation, as we expect from basic thermodynamic arguments. We also note that the off-diagonal terms are identically zero, implying that the quasiparticle and phonon fluctuations are independent. This is somewhat surprising since, as we will see later, the presence of the phonons does significantly modify the spectrum of the quasiparticle fluctuations.

Experimentally, we can only measure the spectrum of the quasiparticle fluctuations, so we will only calculate that spectrum. Using Eq. (13) and quite a bit of algebra, we obtain the quasiparticle spectrum

$$S(\omega) \equiv G_{11}(\omega) = \frac{2\alpha_1\tau_1 N^0}{1 + (\omega\tau_1)^2} + \frac{2\alpha_2\tau_2 N^0}{1 + (\omega\tau_2)^2}, \quad (20)$$

where

$$\alpha_1 = 2 \frac{\tau_1 - \tau_{\text{es}}}{\tau_1 - \tau_2}, \quad \alpha_2 = 2 \frac{\tau_{\text{es}} - \tau_2}{\tau_1 - \tau_2}$$

and  $\gamma_{1,2} = 1/\tau_{1,2}$  are the eigenvalues of  $\mathbf{M}$  and  $\tau_{\text{es}} = 1/\Gamma_{\text{es}}$ . It is straightforward to show that if we integrate  $S(\omega)$  over all  $\omega$  we recover  $N^0$  for the variance. This expression is completely general. However, in the limit  $\Gamma_R \ll \Gamma_B + \Gamma_{\text{es}}$  we can simplify the eigenvalues of  $\mathbf{M}$  to  $\tau_1 = 1/\Gamma_R^*$  and  $\tau_2 = (\Gamma_{\text{es}} + \Gamma_B)^{-1}$ , where  $\Gamma_R^*$  is defined in Eq. (18). In this case, one time constant basically corresponds to the effective quasiparticle lifetime and one corresponds to the phonon lifetime. We can then interpret the first term of Eq. (20) as ‘‘intrinsic’’ quasiparticle fluctuations and the second term as phonon-driven fluctuations.

In many experimental situations at low temperatures, we expect that  $\Gamma_R < \Gamma_B + \Gamma_{\text{es}}$  by several orders of magnitude (Sec. IV B). In this extreme limit, we have that  $\alpha_1 \approx 2$  and  $\alpha_2 \approx 0$ . This gives us a simplified expression for the spectrum

$$S(\omega) \approx \frac{4\tau_R^* N^0}{1 + (\omega\tau_R^*)^2}.$$

If we compare this simplified  $S(\omega)$  with the one-variable result found in Eq. (7), we see that this power spectrum could have been obtained from a simpler one-variable master equation assuming effective generation and recombination parameters

$$r(N) = \frac{1}{2} \frac{R}{F_\omega \text{vol}} N^2, \quad g(N) = r(N^0),$$

where the generation parameter  $g(N)$  is just a constant equal to the equilibrium recombination rate. This simplification is not general, but it is possible in samples where the quasipar-

ticle and phonon time scales are widely separated. Basically, the quasiparticle system cannot respond to the fast phonon fluctuations and is only affected by the average number of phonons.

### C. Extrinsic quasiparticle fluctuations

For our second case, we consider extrinsic quasiparticle fluctuations where we allow quasiparticles to be lost to processes other than recombination. In particular, we consider additional processes that change the number of quasiparticles by 1. There are many physical examples of this kind of process including trapping into material defects,<sup>12</sup> diffusion, trapping into normal-metal regions induced by fluxons,<sup>13</sup> and trapping into external normal-metal ‘‘sinks.’’<sup>14</sup> The multi-variable theory presented here could be applied to a system with an arbitrary number of these extrinsic loss processes. However, we will develop the theory for only one extrinsic loss process in addition to intrinsic recombination. If we were to consider such a system fully, including phonons, we would have a four level system described by a three variable master equation. However, we saw in the previous section that in many relevant experimental systems the effect of the phonons reduces to simply modifying the recombination constant. We therefore consider only a three level system with an effective recombination constant  $R^*$ .

Our three levels are (1) the number of free quasiparticles, (2) the number of trapped quasiparticles, and (3) the number of pairs. The levels are described by the occupation numbers  $N$ ,  $N_t$ , and  $N_p$ , respectively, and we take  $N$  and  $N_t$  to be independent. We assume the allowed transition parameters are  $p_{12} = \Gamma_t N$ ,  $p_{21} = \Gamma_d N_t$ ,  $p_{13} = R^* N^2 / (2 \text{vol})$ , and  $p_{31} = p_{13}^0 = R^* (N^0)^2 / (2 \text{vol})$  where  $\Gamma_t$  is the trapping rate and  $\Gamma_d$  is the detrapping rate. We also write the shot sizes as  $\delta n_{12} = 1$ ,  $\delta n_{21} = 1$ ,  $\delta n_{13} = 2$ , and  $\delta n_{31} = 1$ . We have made some implicit assumptions in writing these transition parameters. First, we have assumed that we are working at low temperatures such that the number of pairs is much greater than the number of quasiparticles. Second, we have assumed that our traps are far from being saturated, such that the transition parameters do not depend on the number of available trap states. With these parameters and assumptions we can apply Eqs. (9) and (10) to find

$$\mathbf{M} = \begin{pmatrix} \Gamma_R^* + \Gamma_t & -\Gamma_d \\ -\Gamma_t & \Gamma_d \end{pmatrix},$$

$$\mathbf{B} = \begin{pmatrix} 2(\Gamma_R^* + \Gamma_t)N^0 & -(\Gamma_t N^0 + \Gamma_d N_t^0) \\ -(\Gamma_t N^0 + \Gamma_d N_t^0) & 2\Gamma_d N_t^0 \end{pmatrix}$$

$$= 2N^0 \begin{pmatrix} \Gamma_R^* + \Gamma_t & -\Gamma_t \\ -\Gamma_t & \Gamma_t \end{pmatrix},$$

where  $\Gamma_R^* = 2R^* N^0 / \text{vol}$ . In simplifying  $\mathbf{B}$ , we have applied the principle of detailed balance, i.e., assumed  $p_{21}^0 = p_{12}^0$  and  $p_{31}^0 = p_{13}^0$ . As discussed earlier, this is always valid for a system in thermodynamic equilibrium, but it must also be true for our system in steady-state or  $N_t$  and  $N_p$  would not have

well-defined steady-state values. We can therefore use the quasiequilibrium result (11) to calculate the covariance matrix

$$\sigma^2 = \begin{pmatrix} N^0 & 0 \\ 0 & N_t^0 \end{pmatrix} = \begin{pmatrix} N^0 & 0 \\ 0 & \frac{\Gamma_t}{\Gamma_d} N^0 \end{pmatrix}.$$

We see that, even though the quasiparticles are now connected to more than one level, the variance of their fluctuation is still simply  $N^0$ .

We could now calculate the general power spectra of this model, but the equations are not particularly illuminating. Instead, we will further simplify the model to the experimentally interesting case where trapping and detrapping are the faster processes. Specifically, we will assume that  $\Gamma_t + \Gamma_d \gg \Gamma_R^*$ . In this limit, the eigenvalues of  $\mathbf{M}$  are

$$\gamma_1 = \frac{\Gamma_d}{\Gamma_d + \Gamma_t} \Gamma_R^*, \quad \gamma_2 = \Gamma_d + \Gamma_t.$$

The spectrum of the quasiparticle fluctuations is then

$$S(\omega) \equiv G_{11}(\omega) = \frac{S_1}{1 + (\omega\tau_1)^2} + \frac{S_2}{1 + (\omega\tau_2)^2}, \quad (21)$$

where

$$S_1 = 4N^0\tau_1 \left[ \frac{\Gamma_d - \Gamma_R^*}{\gamma_2 - \gamma_1} \right] \approx 4N^0\tau_1 \frac{\Gamma_d}{\Gamma_d + \Gamma_t}$$

and

$$S_2 = 4N^0\Gamma_t(\tau_2)^2 \left[ \frac{\gamma_2 + \Gamma_R^*}{\gamma_2 - \gamma_1} \right] \approx 4N^0\tau_2 \frac{\Gamma_t}{\Gamma_d + \Gamma_t},$$

where the final simplification of  $S_1$  and  $S_2$  represent extreme limits. As before, the spectrum is the sum of two Lorentzians each with a bandwidth determined by the eigenvalues of  $\mathbf{M}$ . The relative weight of each Lorentzian depends on the depth of the traps. We call the traps “deep” if  $\Gamma_t > \Gamma_d$ , meaning that once a quasiparticle is trapped it takes a relatively long time for it to escape. Conversely, we call the traps “shallow” if  $\Gamma_d > \Gamma_t$ , meaning that quasiparticles escape relatively quickly. For very deep traps,  $\gamma_2 \approx \Gamma_t$  and  $S_2$  dominates  $S_1$ , such that

$$S_{\text{deep}}(\omega) \approx \frac{4\tau_t N^0}{1 + (\omega\tau_t)^2},$$

where  $\tau_t = 1/\Gamma_t$ . This is the result we would expect for a two-level system where quasiparticles can be lost only to traps. For very shallow traps, we instead find

$$S_{\text{shallow}}(\omega) \approx \frac{4\tau_R^* N^0}{1 + (\omega\tau_R^*)^2},$$

which is the result we expect for quasiparticles in the presence of recombination only. Equation (21) varies smoothly between these two cases and it is easy to show that the integral of the power spectrum is  $N^0$  for any trap depth.

## IV. DISCUSSION

### A. Theoretical connections

We note that comparing the two rate matrices (17) and (19),  $\Gamma = \mathbf{M}$  for the intrinsic fluctuation problem. As we have seen, the eigenvalues of  $\mathbf{M}$  determine the spectrum of the fluctuations. On the other hand, the eigenvalues of  $\Gamma$  determine the time constants of the dynamical response to small perturbations. The fact that these two matrices are equal implies that the timescales of the dynamical response and the fluctuations are the same. We will now take some time to explore the generality of this connection beyond the specific example of intrinsic fluctuations.

When we write rate equations such as (14)–(16) we are making some implicit approximations. First, we approximate the occupation numbers of the levels, such as  $N$  and  $N_\omega$ , as continuous variables, when they are in fact discrete variables. Second, we approximate the discrete and random transitions between levels as continuous and deterministic “flows.” To understand the implications of these approximations, we start by deriving a differential equation for the expectation values of the level occupations from the master equation (8). We find the following system of equations:

$$\frac{\partial}{\partial t} \langle N_i \rangle = \sum_{j \neq i} \delta n_{ij} (\langle p_{ji}(N_j) \rangle - \langle p_{ij}(N_i) \rangle), \quad (22)$$

where the indices  $i$  and  $j$  run over all levels. We can compare this equation to a general expression for the rate equations [similar to Eq. (6)], which is

$$\frac{\partial}{\partial t} N_i = \sum_j \delta n_{ij} [p_{ji}(N_j) - p_{ij}(N_i)]. \quad (23)$$

The only apparent difference is that we have dropped the expectation value brackets from the second system of equations. However, we must also keep in mind the subtle difference that the first equation is an exact differential equation for the continuous expectation value of a discrete variable. The second equation is only approximate, for the reasons mentioned above.

However, in the special case where the  $\{p_{ij}(N_i)\}$  are all linear functions of the occupation numbers  $\{N_i\}$ , we have that  $\langle p_{ij}(N_i) \rangle = p_{ij}(\langle N_i \rangle)$  and we can actually interpret the rate equations (23) as exact equations for the expectation values. In many physical systems, although, the transition rates are at least quadratic in the occupation numbers, such that,  $p_{ij} \sim N_i^2$  or  $p_{ij} \sim N_i N_j$ . In this case, we have, for example, that

$$\langle p_{ij} \rangle \sim \langle N_i^2 \rangle = \langle N_i \rangle^2 + \langle \Delta N_i^2 \rangle.$$

Thus, in the case of quadratic transition rates we must interpret the full nonlinear rate equations as only approximate equations for the expectation values, ignoring terms of order the variance of the occupation number. In general though, we expect that  $\langle \Delta N_i^2 \rangle \sim \langle N_i \rangle$  and we can say that neglecting the variance terms is a valid approximation to order  $\mathcal{O}(1/N)$ . In



other words, for a large system the rate equations actually describe the expectation values of the occupation numbers, to good approximation.

We can develop this idea a little further. If we take Eq. (22) and Taylor expand the transition probabilities to first order we get the following equation for small variations:

$$\begin{aligned} \frac{\partial}{\partial t} \langle \Delta N_i \rangle &= \sum_{j \neq i} \delta n_{ij} \left[ \sum_k \frac{\partial p_{ji}}{\partial N_k} \langle \Delta N_k \rangle \right. \\ &\quad \left. - \sum_k \frac{\partial p_{ij}}{\partial N_k} \langle \Delta N_k \rangle \right] \Big|_{\{N_k\}=\{N_k^0\}} \\ &= \sum_k M_{ik} \langle \Delta N_k \rangle, \end{aligned}$$

where  $M_{ik}$  are the elements of the matrix  $\mathbf{M}$  defined in Eq. (9). If we follow the same procedure for the rate equations, and we find that the linearized rate equations

$$\begin{aligned} \frac{\partial}{\partial t} \Delta N_i &= \sum_{j \neq i} \delta n_{ij} \left[ \sum_k \frac{\partial p_{ji}}{\partial N_k} \Delta N_k - \sum_k \frac{\partial p_{ij}}{\partial N_k} \Delta N_k \right] \Big|_{\{N_k\}=\{N_k^0\}} \\ &\equiv \sum_k \Gamma_{ik} \Delta N_k, \end{aligned}$$

where we have defined the linearized rate matrix  $\Gamma$ . We see, in general now, that  $\Gamma = \mathbf{M}$  and that we can interpret the linearized rate equations as equations for the expectation values around their steady-state values. This result is the general connection between fluctuations and dynamics we were seeking. It says that the timescales measured from dynamic perturbations and from steady-state fluctuations must be the same. We can also view this result as a statistical fluctuation-dissipation theorem for our system. In fact, in equilibrium, we can derive the fluctuations of our system in a thermodynamic framework using the more conventional fluctuation-dissipation theorem.<sup>7,15</sup>

### B. Experimental connections

In a previous Letter, we presented experimental verification of our theory by measuring intrinsic quasiparticle number fluctuations in an Al box.<sup>5</sup> The box was formed by a volume  $\text{vol} = 100 \mu\text{m}^3$ , of thin-film superconducting Al. Two sides of the box were contacted by superconducting Ta leads. The Ta leads allow electrical contact to the box through the Cooper pair system, while still confining quasiparticles in the Al (Fig. 3). Thermal quasiparticles in the Al cannot enter the Ta because the energy difference between the superconducting energy gap of Ta ( $\Delta_{\text{Ta}} = 700 \mu\text{eV}$ ) and the energy gap of Al ( $\Delta_{\text{Al}} = 180 \mu\text{eV}$ ) is much greater than  $k_B T \approx 20 - 30 \mu\text{eV}$  and confines the quasiparticles. There are no thermal quasiparticles in the Ta at the temperatures used.

The number of quasiparticles in the box in thermal equilibrium is

$$N(T) = D(\varepsilon_F) \text{vol} \sqrt{2\pi\Delta_{\text{Al}}k_B T} \exp\left(\frac{\Delta_{\text{Al}}}{k_B T}\right),$$

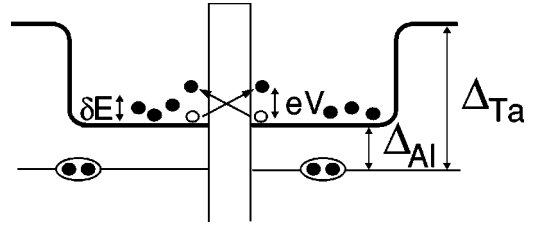


FIG. 3. Energy band diagram of the Al box in a modified excitation representation. The circles represent quasiparticles. Tunneling is shown as diagonal transitions across the barrier, indicating that quasiparticles gain (lose) energy as they are accelerated (decelerated) by the bias voltage. Quasiparticles are confined in both Al electrodes by high gap Ta. At high bias voltage, only electron tunnel is allowed from left to right and only hole tunneling is allowed from right to left. This hole process is known as backtunneling. The two processes allow a single quasiparticle to circulate, tunneling multiple times.

where  $D(\varepsilon_F)$  is the electron density of states at the Fermi energy. In our measurements  $k_B T \ll \Delta_{\text{Al}}$ , so the Fermi gas of quasiparticles is nondegenerate. In particular, the density of quasiparticles is about  $10^{-8}$  that of conduction electrons and the occupation probability of each quasiparticle state is less than  $10^{-3}$ , much smaller than in the normal state at the Fermi energy.

We measure the number of the quasiparticles in the gas by dividing the box with a tunnel barrier and measuring the current through the tunnel barrier. At large bias, there is a simple connection between the number of quasiparticles in the box and the current, given by the next equation. In Fig. 3, we show quasiparticles distributed in an energy range  $\delta E$  in the Al. (For a thermal distribution,  $\delta E$  is a few times  $k_B T$ .) The ovals represent Cooper pairs at the Fermi energy. Each quasiparticle is a quantum superposition of electron and hole. Biased at a voltage  $eV > \delta E$ , a quasiparticle can only tunnel from left to right as an electron, gaining energy  $eV$ . It cannot tunnel from left to right as a hole, because it would lose energy  $eV$  and tunnel into the gap on the right side. Similarly, a quasiparticles can only tunnel from right to left as a hole (through a process called backtunneling).<sup>16</sup> Thus, for  $eV > \delta E$ , tunneling events from left to right and from right to left transfer a charge in the same direction and the associated currents add. The time-dependent current is then given by

$$I(t) = e \left( \frac{N_l(t)}{\tau_{\text{tun}}} + \frac{N_r(t)}{\tau_{\text{tun}}} \right) = e \frac{N(t)}{\tau_{\text{tun}}},$$

where  $N_l$  and  $N_r$  are the numbers of quasiparticles in the left and right side and  $\tau_{\text{tun}}$  is the tunnel time.<sup>17</sup> In writing this equation, we have assumed that any variations in  $N(t)$  happen on a time scale  $\tau \gg \tau_{\text{tun}}$ . As we will show later, the time scale of the fluctuations in the box meets this condition.

It is a good approximation to treat the two halves as one quasiparticle system if the halves are strongly coupled. The condition for strong coupling is  $\tau_R^* \gg \tau_{\text{tun}}$ , where  $\tau_R^*$  is the effective recombination time for a quasiparticle. If this condition is met, a typical quasiparticle tunnels many times before it recombines, and thus can interact with quasiparticles

in both halves of the box. In our measurements,  $n = \tau_R^*/\tau_{\text{tun}}$  is between 10 and 50. The fact that a quasiparticle in a superconductor is a superposition of electron and hole allows it to tunnel back and forth multiple times.<sup>18</sup>

We also directly measured the recombination time of quasiparticles in the box with single-photon absorption experiments.<sup>19</sup> A single photon from the mercury emission line at 4.89 eV (256 nm) was absorbed in one Ta lead, producing about 4000 quasiparticles. These quasiparticles diffuse to the Al where they can emit phonons and drop down in energy, becoming trapped. These trapped quasiparticles are a small perturbation to the  $N^0 \sim 10^5$  steady-state quasiparticles in the Al box. The trapped quasiparticles circulate, tunneling and backtunneling, until they are lost to recombination with a thermal quasiparticle. This produces a current pulse that decays exponentially on a time scale of the effective recombination time  $\tau_R^*$ .

In thin-film Al electrodes at the temperatures used we expect  $\Gamma_R \approx 10^4 \text{ s}^{-1}$ ,  $\Gamma_{\text{es}} \approx 10^9 \text{ s}^{-1}$ , and  $\Gamma_B \approx 10^{10} \text{ s}^{-1}$  (Ref. 20). Thus, referring to Eq. (20), we expect  $\alpha_1 \approx 2(1 - 10^{-5})$  and  $\alpha_2 \approx 2(10^{-5})$ . This gives us a simplified expression for the spectrum

$$S(\omega) \approx \frac{4\tau_R^* N^0}{1 + (\omega\tau_R^*)^2}.$$

This result contains three important predictions. First, it predicts that the power spectrum of the fluctuations should be Lorentzian. Second, it predicts the temperature dependence of the bandwidth of the noise. Basically, the bandwidth is proportional to the average number of quasiparticles, since  $1/\tau_R^* \sim N^0$ .  $N^0$  is an exponential function of  $1/T$  well below  $T_c$ , so, we predict that the bandwidth is also an exponential function of  $1/T$  in our temperature range. The theory also predicts how the low-frequency magnitude of the noise  $S(\omega=0)$ , changes as a function of temperature. All factors in the magnitude of  $S(\omega=0)$  are approximately independent of temperature except  $N^0$  and  $\tau_R^*$ . However,  $\tau_R^*$  only changes with temperature because the number of quasiparticles changes. Specifically,  $\tau_R^*(T) \sim 1/N^0(T)$ . Thus, the product  $N^0\tau_R^*$  is constant and, therefore,  $S(\omega=0)$  should be independent of temperature.

Our measurements showed good agreement with all of these predictions. First, we confirmed that the quasiparticle fluctuations had a Lorentzian form. We also confirmed that the characteristic time of the fluctuations was in fact  $\tau_R^*$ , over a range of temperatures, by comparing the noise measurements to the direct measurement of  $\tau_R^*$  given by photon excitation. We were also able to indirectly confirm the temperature dependence of  $S(\omega=0)$ . We were not able to directly confirm the temperature dependence because the devices were heated by the bias power and, therefore, had an effective temperature higher than the bath temperature. Still, we demonstrated that the magnitude of the noise was independent of the quasiparticle density, which is a measure of the effective temperature.

In superconducting systems, at least in principle, the fundamental time scale of electron-phonon interactions, known

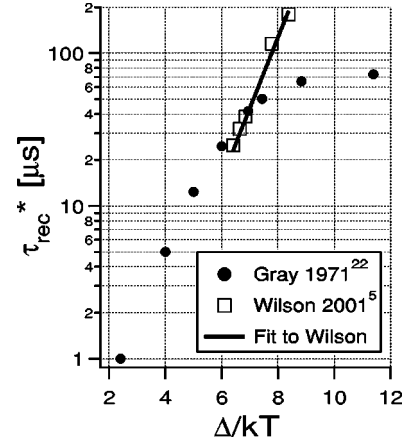


FIG. 4. Comparison of quasiparticle lifetime measurements described here to previous measurements by Gray (Ref. 22). Measurements by Gray were on Al on sapphire with  $\Delta = 195 \mu\text{V}$ . Our films are on  $\text{SiO}_2$  with  $\Delta = 180 \mu\text{V}$ . The solid line shows the theoretical scaling of the lifetime with the BCS number of quasiparticles for our value of  $\Delta$ . Our data show the lifetime following the theoretical dependence to lower temperature (Ref. 5).

as  $\tau_0$ , can be inferred from measurements of quasiparticle-quasiparticle recombination. (The parameter  $\tau_0$  is material dependent and its value has been predicted for a variety of metals.<sup>20</sup>) In particular, for a pair of quasiparticles at the gap edge, the expression for the recombination constant is

$$R = \left( \frac{2\Delta}{k_B T_c} \right)^3 \frac{1}{2\Delta D(\epsilon_F) \tau_0}, \quad (24)$$

where  $\Delta$  is the superconducting energy gap,  $T_c$  is the superconducting transition temperature, and  $D(\epsilon_F)$  is the electron density of states at the Fermi energy. However, phonon trapping complicates the extraction of  $\tau_0$  from recombination measurements at temperatures much less than  $T_c$ . In fact, in the limit of strong phonon trapping, the measured recombination rate  $\Gamma_R^*$  becomes

$$\Gamma_R^* = 2 \frac{\Gamma_R}{\Gamma_B} \Gamma_{\text{es}} \sim \Gamma_{\text{es}}$$

because the pair-breaking rate  $\Gamma_B$  is also proportional to  $1/\tau_0$ . Thus, measurements of  $\Gamma_R^*$  in the presence of strong phonon trapping have no dependence on  $\tau_0$ .

As described above, we have used both fluctuations and photoexcitation to measure  $\Gamma_R^*$  in Al. If we ignore phonon trapping for the moment and insert our measured value of  $R$  into Eq. (24), we extract a tentative value for  $\tau_0$  of  $1.65 \mu\text{s}$ . Numerous other measurements of  $\tau_0$  in Al by various methods find values of order 100 ns.<sup>11,21</sup> This discrepancy suggests that our measurements are, in fact, in the limit of strong phonon trapping, so that they do not represent a direct measurement of  $\tau_0$ . Our measurements do, however, confirm that the quasiparticle recombination rate is proportional to the quasiparticle density at lower temperatures and longer recombination times than previous experiments. In Fig. 4, we compare the recombination time measured by us to previous experiments and to theory. The previous measurements

showed recombination times that begin to deviate from the expected dependence at  $T \approx 400$  mK and  $\tau_R^* \approx 20 \mu\text{s}$  and completely saturate at a maximum value of  $\tau_R^* \approx 80 \mu\text{s}$  below  $T \approx 300$  mK.<sup>22</sup> Quasiparticle loss into normal-metal regions created by trapped flux was proposed as the explanation for the deviation from theory in those measurements, although this explanation was not experimentally confirmed. Therefore, our measurements extend the range over which the basic physics of recombination has been verified in Al.

While quasiparticle number fluctuations may be helpful in studying the microscopic dynamics of superconductors, they are also a source of noise in superconducting electronic devices. We have discussed in detail how they can limit the performance of single-photon spectrometers based on superconducting tunnel junctions.<sup>23</sup> In addition, quasiparticle fluctuations may be an important source of noise, and therefore decoherence, in superconducting quantum bits (qubits). The majority of solid-state systems that have been used to demonstrate coherent quantum manipulation of a single qubit have involved superconductors.<sup>3</sup> All of these measurements have been performed at very low temperatures ( $T/T_c \sim 0.01$ ), where there would be essentially zero quasiparticles in equilibrium. However, all of the readout schemes in these experiments produce nonequilibrium quasiparticles, which can accumulate in the qubits, leading to a steady-state den-

sity of quasiparticles. At least one experiment has directly demonstrated the impact of this quasiparticle background on the measured coherence times.<sup>24</sup> Understanding the effect of quasiparticle fluctuations on coherence may therefore be important for the development of quantum bits.

In conclusion, we have developed a general theory of quasiparticle number fluctuations in superconductors. We applied this general theory to the problem of intrinsic quasiparticle fluctuations related to generation and recombination. The validity of these results have been demonstrated in previous experimental work. We have also applied the theory to an example of extrinsic quasiparticle fluctuations where quasiparticles are also lost to traps. We conclude that studies of quasiparticle fluctuations provide a useful probe of microscopic dynamics and are also important for the understanding of noise in superconducting devices.

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