Time-Resolved Measurements of Thermodynamic Fluctuations of the Particle Number in a Nondegenerate Fermi Gas

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(Received 12 February 2001; published 20 July 2001)

We report on time-resolved measurements of thermodynamic fluctuations in the number of particles in a nondegenerate Fermi gas. The gas is comprised of thermal quasiparticles, confined in a superconducting Al box by large-gap Ta leads. The average number of quasiparticles is about 10^5. This number fluctuates due to quasiparticle generation and recombination. The number is measured from the tunneling current through a barrier that bisects the box. The recombination time is independently measured by single-photon excitation and agrees with the frequency dependence of the fluctuations.

DOI: 10.1103/PhysRevLett.87.067004 PACS numbers: 74.40.+k, 05.40.–a, 05.70.Ln, 85.25.Oj

Statistical mechanics elucidates the microscopic origins of the laws of thermodynamics, connecting the thermodynamic quantities of a system to ensemble averages of microscopic quantities. Thermodynamics is successful because the fluctuations of thermodynamic quantities are relatively small in macroscopic systems. However, fluctuations of thermodynamic quantities can be significant in "small" systems. For example, consider a small container filled with a gas that can exchange energy and particles with a reservoir. If there are N_0 particles on average, the rms amplitude of the fluctuations in the number is exactly $(N_0)^{1/2}$ [1]. Thus, the relative fluctuations are large if N_0 is small. The frequency spectrum of the fluctuations is determined by the dynamics of the specific system being studied. In particular, if the characteristic time that a particle spends in the gas before returning to the reservoir is τ, then the spectrum of the fluctuations extends to a characteristic frequency $\sim 1/\tau$.

In this Letter we present time-resolved measurements of thermodynamic fluctuations of the particle number in a nondegenerate Fermi gas comprised of quasiparticles in a small superconducting Al box. The quasiparticle gas is coupled to a particle and energy reservoir formed by the Cooper pair and phonon systems. We present a model that connects the fluctuations to quasiparticle generation and recombination. The dynamic time scale of the quasiparticle recombination process is independently measured using single-photon absorption to perturb the quasiparticle number. The time scale of recombination agrees well with the time scale of the fluctuations, confirming our model. To our knowledge, this is the first such measurement in a superconducting system.

The box is formed by a volume, vol $= 100\ \mu\text{m}^3$, of thin-film superconducting Al. Two sides of the box are 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. Thermal quasiparticles in the Al cannot enter the Ta because the energy difference between the superconducting energy gap of Ta ($\Delta_{\text{Ta}} = 700\ \mu$eV) and the energy gap of Al ($\Delta_{\text{Al}} = 180\ \mu$eV) is much greater than $k_BT \sim 20$–$30\ \mu$eV (see Fig. 1). There are no thermal quasiparticles in the Ta at the temperatures used. The number of quasiparticles in the box in thermal equilibrium is

$$N(T) = D(\varepsilon_F)\text{vol}\sqrt{2\pi\Delta_{\text{Al}}k_BT}\exp\left(-\frac{\Delta_{\text{Al}}}{k_BT}\right), \quad (1)$$

where $D(\varepsilon_F)$ is the electron density of states at the Fermi energy. In our measurements $k_BT \ll \Delta_{\text{Al}}$, so the Fermi gas is nondegenerate with a density about 10^{-8} that of the normal-state electrons.

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 voltage there is a simple connection between the number of quasiparticles in the box and the current. In the inset of Fig. 1 we show quasiparticles distributed in an energy range δE in the Al. (For a thermal distribution, δE is a few times

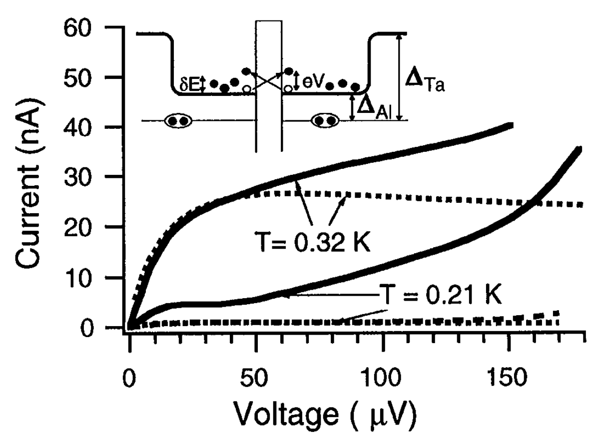

FIG. 1. Current-voltage characteristics of the tunnel junction that divides the Al box. The solid lines are I-V curves from device 1 that show the effect of heating. The dashed line is an I-V curve from a similar junction, device 2, with no Ta contact on the right side and no evidence of heating effects. The dotted lines are BCS predictions. The low temperature BCS curve and the curve from device 2 overlap at low voltage. The inset is an energy band diagram of device 1 in the excitation representation.

k_BT.) 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 quasiparticle in the range δE on the right can only tunnel to the left as a hole (through a process called backtunneling) [2]. Thus, for eV $> \delta E$, tunneling events from left to right and from right to left transfer a negative charge from left to right. The time-dependent current is then given by

$$I(t) = e\left(\frac{N_l(t)}{\tau_{\text{tun}}} + \frac{N_r(t)}{\tau_{\text{tun}}}\right) = e\,\frac{N(t)}{\tau_{\text{tun}}}, \qquad (2)$$

where N_l and N_r are the numbers of quasiparticles in the left and right sides and τ_{tun} is the tunnel time [3]. In writing Eq. (2) 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 gas 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 $\tau_{\text{tun}} \ll \tau_R^*$, where τ_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 [4].

To measure the quasiparticle number with a tunnel barrier, we must use a voltage eV $> \delta E$. The voltage leads to joule heating. Each tunneling event adds eV of energy to a quasiparticle in the Al box. Quasiparticles will emit a phonon after one or more tunneling events, before reaching an energy where they could "climb over" the large-gap Ta leads. Thus, all of the energy added by the bias voltage is dissipated in the Al box, heating it and generating excess quasiparticles. Figure 1 shows current-voltage (I-V) curves from two different devices along with theoretical BCS I-V curves. Device 2 has a Ta lead only on the left side, so hot quasiparticles can diffuse away from the junction on the right side. Its I-V curve represents the behavior of a junction in equilibrium at the bath temperature and is quantitatively consistent with BCS predictions. The current of device 2 also scales with the junction area when compared to the larger junctions we have made with one Ta lead. Device 1, which has Ta leads on both sides and is used in the rest of the measurements in this Letter, shows excess current compared to device 2 [5]. The excess current in device 1 is due to joule heating, as described above.

We directly measure the recombination time of quasiparticles in the box with single-photon absorption experiments [6]. A single photon from the mercury emission line at 4.89 eV (254 nm) is absorbed in the Ta lead on the left of Fig. 1, 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 but are a larger perturbation than typical fluctuations. The trapped quasiparticles circulate, tunneling and backtunneling, until they are lost to recombination with a thermal quasiparticle. This circulation produces a current pulse that decays exponentially on a time scale of the effective recombination time, τ_R^*.

We now develop a model that connects the frequency dependence of the fluctuations to the dynamics of quasiparticle generation and recombination. In a thin-film superconductor, a phonon emitted by a pair of recombining quasiparticles can break another Cooper pair before escaping from the film to the bath [7]. An effective recombination rate, Γ_R^*, then applies for fluctuations and for small perturbations. In the limit $\Gamma_R \ll \Gamma_B + \Gamma_{ES}$, it is

$$\Gamma_R^* = 2\Gamma_R F_\omega^{-1}, \qquad F_\omega = 1 + \frac{\Gamma_B}{\Gamma_{ES}},$$

where Γ_R is the recombination rate, Γ_B is the phonon pair-breaking rate, and Γ_{ES} is the phonon escape rate [8]. Typically, $\Gamma_R^* \ll \Gamma_R$. F_ω is the phonon trapping factor; F_ω^{-1} is the probability that a phonon escapes to the bath.

To treat fluctuations in the 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 of generation-recombination noise in semiconductors [9], except that we generalize the description to allow for transitions that involve an arbitrary number of particles, e.g., two quasiparticles recombining. The Al box is well described by the three level system of Rothwarf and Taylor [7]. The occupants of the levels are quasiparticles in the box, pair-breaking phonons in the box, and pair-breaking phonons in the bath. However, in an Al box with our geometry and our temperature range, we expect $\Gamma_R \sim 10^5\ \text{s}^{-1}$, $\Gamma_{ES} \sim 10^9\ \text{s}^{-1}$, and $\Gamma_B \sim 10^{10}\ \text{s}^{-1}$ [10]. Because the phonon time scales are so much faster than quasiparticle recombination, the quasiparticle fluctuations are affected only by the average number of phonons. This allows us to simplify the model.

We can treat the system as a two level system with effective generation and recombination probabilities [11]. The occupants of the two levels are quasiparticles and Cooper pairs. However, the numbers of quasiparticles and Cooper pairs are not independent; the loss of two quasiparticles corresponds to the gain of one Cooper pair. Thus, we need only to keep track of the total number of quasiparticles in the two halves of the box, $N(t)$. We can then describe our system with a one-variable master equation with three parameters: the effective probability of a recombination event in the box per unit time $r(N)$, the generation probability $g(N)$, and the "shot" size δN. For our system, the effective recombination and generation probabilities per unit time are, respectively, $r(N) = (R/2F_\omega \text{vol})N(t)^2$ and

$g(N) = r(N_0)$, where R is the recombination constant and the generation parameter, $g(N)$, is just a constant equal to the steady-state recombination rate. The shot size is $\delta N = 2$ because quasiparticles are generated and recombine in pairs. We find the following spectral density for the number fluctuations:

$$S_N(\nu) = \frac{4\sigma^2\tau}{1 + (\omega\tau)^2} = \frac{4N_0\tau_R^*}{1 + (\omega\tau_R^*)^2},$$

where

$$\tau = \frac{1}{\delta N[r'(N_0) - g'(N_0)]} = \tau_R^*$$

is the characteristic time of the fluctuations,

$$\sigma^2 = (\delta N)^2 r(N_0)\tau = N_0$$

is the variance of the fluctuations, ν is the frequency, and $\omega = 2\pi\nu$. The primes indicate the derivative with respect to N, and N_0 is the time-average value of $N(t)$. In steady-state $\Gamma_R = RN_0/\mathrm{vol}$, so the time constant of the fluctuations is $\tau_R^* = 1/\Gamma_R^*$.

Equation (2) implies that the fluctuations in the number of quasiparticles will cause fluctuations in the tunneling current with a spectral density

$$S_I(\nu) = \frac{4neI_0}{1 + (\omega\tau_R^*)^2} = \frac{2(\alpha n)eI_0}{1 + (\omega\tau_R^*)^2}, \qquad (3)$$

where I_0 is the steady-state current, $n = \tau_R^*/\tau_{\mathrm{tun}}$ is the average number of times a quasiparticle tunnels before recombining, and we have introduced the parameter α; $\alpha = 2$ in our model. We have written the spectrum in this form to recall the magnitude of $S_I(\nu = 0)$ for Poisson shot noise of a tunneling current I_0, namely, $S_I(\nu = 0) = 2q_{\mathrm{eff}}I_0$, where q_{eff} is the effective charge of the current carriers [12]. Equation (3) looks like a shot noise spectrum with an effective charge $q_{\mathrm{eff}}/e = \alpha n$. Our model predicts $q_{\mathrm{eff}} \sim 100e$ at the lowest temperature. Poisson shot noise arises from the random timing of tunneling events. At high frequency we expect to recover the result $S_I(\nu \gg 1/\tau_R^*) = 2eI_0$ in our junctions.

An additional source of noise could be a fluctuating charge imbalance between the holelike and electronlike quasiparticle branches [2]. In Al away from T_c, these fluctuations can be caused by quasiparticles randomly switching branches upon scattering elastically from an impurity [13]. The branch mixing time in Al is of order $\tau_Q \sim 10^{-8}$ s [13], much faster than either the tunneling time, $\tau_{\mathrm{tun}} \sim 10^{-6}$ s, or the effective recombination time $\tau_R^* \sim 10^{-4}$ s. This means that charge imbalance fluctuations should be averaged out in our measurements. (In addition, τ_Q should be independent of temperature in our operating range, compared to the exponential temperature dependence of τ_R^*.)

We have fabricated and measured Al boxes at Yale [6,14]. We measured a residual resistance ratio of 10 for a typical 200-nm-thick film. Measurements were performed in a two-stage ^{3}He Dewar with a base temperature of 0.21 K. All measurements were made with the tunnel junction biased at $V = 60\ \mu$V. We measured both the exponential fall time of single-photon pulses and the frequency spectrum of the current noise at different temperatures, as shown in Fig. 2. The current pulse shown is the average of 500 single-photon current pulses. The decay is fit with a simple exponential to give one measurement of τ_R^*. The spectrum is the average of fast Fourier transforms of successive noise traces recorded with a digital oscilloscope. We fit a Lorentzian shape such as (3) with a white noise background to give a second measurement of τ_R^*. Before fitting we digitally subtracted known electronic noise sources and removed lines caused by 60 Hz pickup and microphonics [15].

In Fig. 3 we plot both measurements of $\Gamma_R^* = 1/\tau_R^*$ versus the average number of quasiparticles in the box, $N_0(T)$. $N_0(T)$ is inferred from the average current using (2). Each $N_0(T)$ corresponds to a different bath temperature, between 0.21 and 0.32 K. For the lower values of $N_0(T)$, the effective temperature of the quasiparticle gas was significantly higher than the bath temperature (refer to Fig. 1). The values of Γ_R^* inferred from the noise and the single-photon measurements agree, confirming the connection between the noise and the dynamics of recombination. The solid line in Fig. 3 is a least squares fit to the total data set and is *not* constrained to zero intercept. We see that Γ_R^* varies linearly with $N_0(T)$ and that the intercept of the line is approximately zero. This is expected if Γ_R^* is dominated by quasiparticle-quasiparticle recombination, as opposed to loss in traps, etc. [8]. The quality of the fit suggest that despite the fact that at the lowest bath temperature the quasiparticle gas is heated, it is behaving like it is in internal equilibrium at an effective temperature higher than the bath temperature.

The theory also predicts how the low frequency magnitude of the noise, $S_I(\nu = 0)$, should change as a function

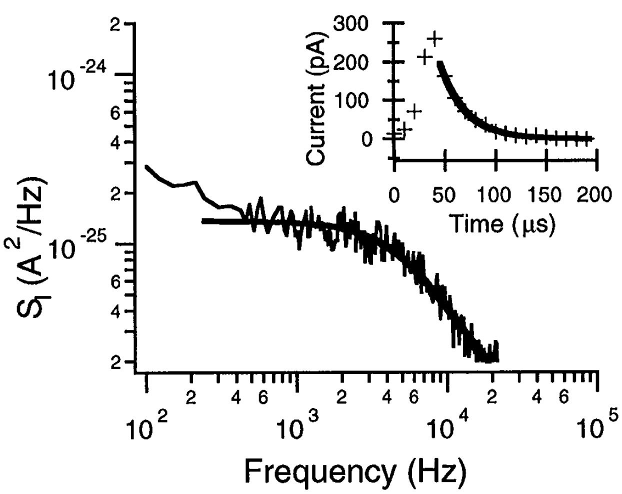

FIG. 2. Spectral density, $S_I(\nu)$, of the tunneling current with a fit to the Lorentzian form (3). The inset is the average of 500 single-photon current pulses showing the decay with a fit to an exponential. For clarity, only a few percent of the data points are displayed. $T = 0.32$ K, $V = 60\ \mu$V.

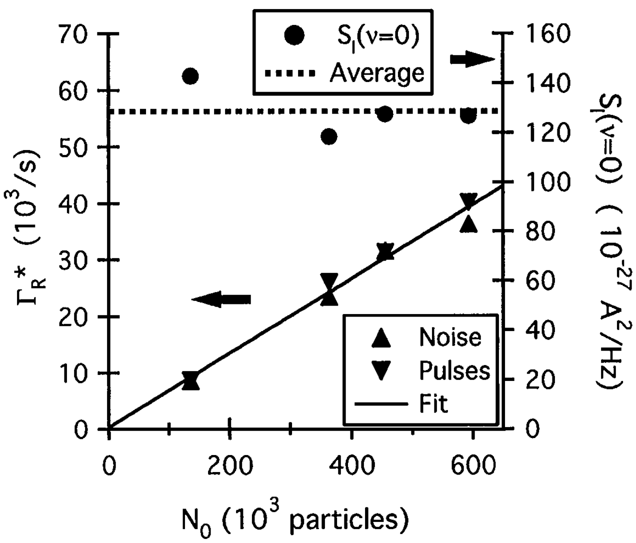

FIG. 3. Data for the effective recombination rate, Γ_R^*, and the low-frequency current spectral density, $S_I(\nu = 0)$. Γ_R^* was determined from the bandwidths of the spectra and the fall times of the current pulses (Fig. 2). The bath temperature ranges from 0.21 to 0.32 K and $V = 60\ \mu$V.

of temperature. Referring to (3), all factors in the magnitude of $S_I(\nu = 0)$ are approximately independent of temperature except I_0 and τ_R^*. I_0 and τ_R^* both change with temperature because the number of quasiparticles changes. Specifically, $I_0 \sim N_0$ and $\tau_R^* \sim 1/N_0$. Thus, the product $I_0\tau_R^*$ is independent of N_0 (temperature) and, therefore, $S_I(\nu = 0)$ should be independent of temperature. The right axis of Fig. 3 shows $S_I(\nu = 0)$, determined by fitting the spectra with (3), plotted versus N_0. We can see that there is *no* significant dependence of $S_I(\nu = 0)$ on N_0. Finally, the theory predicts the absolute magnitude of $S_I(\nu = 0)$, or equivalently the parameter α from (3). We measure a value $\alpha = 1.8 \pm 0.2$. This agrees well with our model prediction of $\alpha = 2$ [16]. The uncertainty quoted is one standard deviation of α and is dominated by the uncertainty in the electron density of states at the Fermi energy, $D(\varepsilon_F)$.

Our work suggests that thermodynamic fluctuations may complicate the study of noise in mesoscopic superconductors [17]. Thermal generation-recombination noise may also be important in superconducting devices. For example, Al boxes are used in single-photon spectrometers [18]. In Ref. [14] we describe how the fluctuations treated above amount to a previously unknown "thermodynamic limit" for superconducting tunnel junction (STJ) spectrometers which use two larger-gap leads to promote backtunneling.

We thank L. Li, M. Devoret, R. Schoelkopf, K. Segall, and A. Kozorezov for useful discussions. This work was supported by NASA-GSFC and NSF/DMR-0072022.

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