Number Fluctuations of Sparse Quasiparticles in a Superconductor

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We have directly measured quasiparticle number fluctuations in a thin film superconducting Al resonator in thermal equilibrium. The spectrum of these fluctuations provides a measure of both the density and the lifetime of the quasiparticles. We observe that the quasiparticle density decreases exponentially with decreasing temperature, as theoretically predicted, but saturates below 160 mK to 25–$55/\mu m^3$. We show that this saturation is consistent with the measured saturation in the quasiparticle lifetime, which also explains similar observations in qubit decoherence times.

In a superconductor the density of unpaired electrons (quasiparticles) should vanish when approaching zero temperature [1]. This crucial property promises long decoherence times for superconducting qubits [2] and long relaxation times for highly sensitive radiation detectors [3]. However, relaxation times for resonators [4,5] and qubit decoherence times [6–8] were shown to saturate at low temperature. Recent modeling [8,9] suggests that nonequilibrium quasiparticles are the main candidate for this saturation, which was tested qualitatively by injecting quasiparticles into a qubit [10]. A direct measurement of the number of quasiparticles and the energy decay rate in equilibrium at low temperatures would provide new insight in superconductivity at low temperatures, crucially needed in the aforementioned fields.

At finite temperature, it follows from thermodynamics that the density of quasiparticles fluctuates around an average value that increases exponentially with temperature [11]. Here we report a measurement of the spectrum of these fluctuations in a single aluminum superconducting film ($T_c = 1.1$ K) in equilibrium, for temperatures from 300 to 100 mK. The number fluctuations show up as fluctuations in the complex conductivity of the film, probed with a microwave resonator. The spectrum of these fluctuations provides a direct measure of the number of quasiparticles in the superconductor. We observe that the quasiparticle density decreases exponentially with decreasing temperature until it saturates at 25–55 μm^{-3} below 160 mK. We prove that the measured saturation of the quasiparticle lifetime to 2.2 ms below 160 mK is consistent with the saturation in quasiparticle density. In addition, our experiment shows that it is possible to reach the fundamental generation-recombination noise limit in detectors based on Al resonators.

In a superconductor in thermal equilibrium, the density of quasiparticles per unit volume is given by

$$n_{qp} = 2N_0\sqrt{2\pi k_B T \Delta}\exp(-\Delta/k_B T), \qquad (1)$$

valid at $k_B T < \Delta$, with N_0 the single spin density of states at the Fermi level (1.72×10^{10} μm^{-3} eV^{-1} for Al), k_B Boltzmann's constant, T the temperature, and Δ the energy gap of the superconductor. Two quasiparticles with opposite spins and momenta can be generated from a Cooper pair by a phonon with an energy larger than the energy gap. When two quasiparticles recombine into a Cooper pair, a phonon is emitted. These processes, schematically depicted in Fig. 1(a), are random processes in equilibrium.
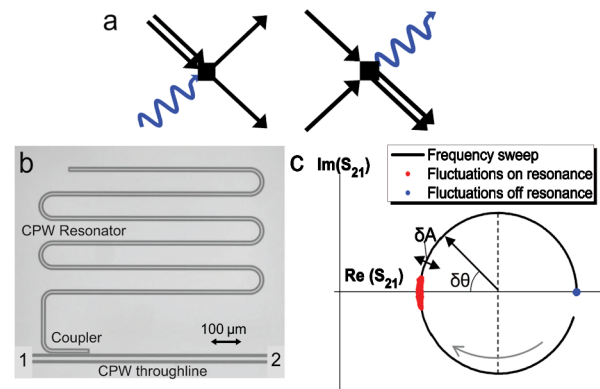

FIG. 1 (color online). (a) Schematic of the process of generation (left) and recombination (right) of quasiparticles in a superconductor. The single arrow symbolizes a quasiparticle, the double arrow a Cooper pair, and the wavy arrow a phonon. (b) A microscope picture of the microwave resonator, 2 times reduced in length for visibility. The open ends set the half-wavelength resonance condition. The coplanar waveguide (CPW) throughline is used for the microwave excitation and readout (from contact 1 to 2) of the resonator. (c) The real and imaginary parts of the complex transmission S_{21} as a function of frequency around the resonant frequency. The gray arrow indicates the direction of increasing frequency. We define a resonator amplitude A and phase θ with respect to the resonance circle center as indicated. In red, the measured fluctuations in S_{21} at the resonant frequency are shown. The fluctuations far off resonance are shown in blue, which are used as a calibration for system noise contributions.

Assuming a thermal distribution of quasiparticles and phonons at low temperature, the average quasiparticle recombination time is given by [12]

$$\tau_r = \frac{\tau_0}{\sqrt{\pi}}\left(\frac{k_B T_c}{2\Delta}\right)^{5/2}\sqrt{\frac{T_c}{T}}e^{\Delta/k_B T} = \frac{\tau_0}{n_{qp}}\frac{N_0(k_B T_c)^3}{2\Delta^2}, \quad (2)$$

where T_c is the critical temperature of the superconductor and τ_0 a material dependent, characteristic electron-phonon interaction time. Equations (1) and (2) predict a very low quasiparticle density and consequently a very long quasiparticle lifetime at temperatures $T < T_c/10$.

The process of random generation and recombination of charge carriers is a well-studied phenomenon in solid state physics, in particular in semiconductors, but has hardly been studied in superconductors. In one earlier experiment, this generation-recombination noise was identified in the current fluctuations through a tunnel barrier connected to a small Al quasiparticle box [13], although only down to an intermediate 210 mK. The general theory of quasiparticle number fluctuations in a superconductor [11,13] connects the frequency dependence of the fluctuations to the microscopic dynamics of quasiparticle generation and recombination. The dominant time scale of these processes is the recombination time of a quasiparticle (τ_r is about 1 ms in an Al film [4]), because the phonon pair breaking time and the phonon escape time are both much shorter, about 10^{-10} s based on a film thickness of 40 nm [12,14]. In thermal equilibrium, the generation and recombination rates are equal and the variance of the random number fluctuations $\sigma^2 = \langle\delta N_{qp}^2\rangle = N_{qp} = n_{qp}V$, with V the volume of the system. The power spectral density of these fluctuations shows a Lorentzian spectrum, given by

$$S_N(\omega) = \frac{4N_{qp}\tau_r}{1 + (\omega\tau_r)^2}, \quad (3)$$

with ω the angular frequency. Equations (1) and (2) show that the product $N_{qp}\tau_r$ is constant over temperature, whereas the total integrated power spectral density increases exponentially with temperature. This is because the bandwidth of the fluctuations increases exponentially with temperature as well, as it scales with $1/\tau_r^2$. We emphasize that this property is unique for quasiparticle generation-recombination noise in a superconductor.

We measure the quasiparticle number fluctuations using a high-quality microwave resonator. The high frequency response of the superconductor is controlled by the quasiparticle density through the complex conductivity $\sigma_1 - i\sigma_2$. The real part, σ_1, is resistive and denotes the conductivity by quasiparticles. The imaginary part, σ_2, is inductive and due to the superconducting condensate [15]. Quasiparticle number fluctuations will show up as fluctuations in the complex conductivity. To measure the complex conductivity, a 40 nm thick Al film was patterned into microwave resonators. The film was sputter deposited onto a C-plane sapphire substrate. The critical

temperature is 1.11 K, from which the energy gap $\Delta = 1.76k_B T_c = 168$ μeV. The low temperature resistivity $\rho = 0.8$ $\mu\Omega$ cm and the residual resistance ratio RRR = 5.2. The film was patterned by wet etching into distributed, half-wavelength, coplanar waveguide resonators, with a defined central linewidth of 3.0 μm and gaps of 2.0 μm wide [Fig. 1(b)]. The resonator under consideration shows its lowest order resonance at 6.619 24 GHz and has a central strip volume of 1.0×10^3 μm^3. The resonance curve at 100 mK shows a coupling limited quality factor of 3.87×10^4. The samples are cooled in a pulse tube precooled adiabatic demagnetization refrigerator. The cold stage is surrounded by a superconducting magnetic shield inside a cryoperm shield. Special care has been taken to make the setup light tight, such that no excess quasiparticles are created by stray light. The sample is mounted inside a light-tight holder which itself is placed inside another light-tight box (also at base temperature), to prevent radiation leaking in via the coax cable connectors. Radiation absorber, consisting of carbon black, epoxy, and SiC grains is placed inside both the sample holder and the outer box. The outer box is equipped with special coax cable filters that attenuate all frequencies above 10 GHz exponentially (see Ref. [16]). The system is proven to be light tight by measuring the quasiparticle lifetime as a function of the 4 K-stage temperature while keeping the sample at base temperature. Within the measurement accuracy (15%), there was no change in the lifetime, indicating that the stray-light power at the chip is negligible. For the lifetime measurements the sample is illuminated with a short pulse of light from a GaAsP light-emitting diode (see Ref. [4]), fiber coupled to the sample via a 0.15 mm waveguide (a 1 THz high pass filter) to filter pair breaking radiation from the 4 K environment [17].

The complex transmission of the microwave circuit is measured with a quadrature mixer and traces out a circle in the complex plane. The microwave signal is amplified at 4 K with a high electron mobility transistor (HEMT) amplifier and with a room temperature amplifier, before it is mixed with a copy of the original signal [3,18]. We define a resonator amplitude and phase with respect to the resonance circle, as depicted in Fig. 1(c). The resonator amplitude predominantly responds to changes in σ_1 [19]. The responsivity of the resonator amplitude to quasiparticles $dA/dN_{qp} = -2\alpha Q\kappa/V$, with Q the quality factor of the resonator, α the fraction of kinetic inductance over the total inductance, and $\kappa = \frac{\delta\sigma_1/\sigma_2}{\delta n_{qp}}$, which depends only weakly on temperature. The amplitude responsivity was determined experimentally as described in Ref. [18]. dA/dN_{qp} is measured to be almost temperature independent. For similar resonators it is known that the sensitivity in phase is limited by two-level fluctuators [20,21] and that the sensitivity in amplitude is up to a factor of 10 better, limited by the HEMT amplifier [18].

The power spectral density due to quasiparticle number fluctuations in the resonator amplitude is given by

$$S_A(\omega) = S_N(\omega) \frac{(dA/dN_{qp})^2}{1 + (\omega\tau_{res})^2}, \qquad (4)$$

where τ_{res} is the resonator ring time given by $\tau_{res} = \frac{Q}{\pi f_0}$. In this experiment $\tau_r \gg \tau_{res} \approx 2\ \mu s$, meaning that the roll-off in the noise spectrum will be determined solely by τ_r. Using Eqs. (1)–(4), with $\tau_0 = 438$ ns [12] and a measured $dA/dN_{qp} = 5.0 \times 10^{-7}$, we expect $S_A = -99.3$ dBc/Hz. This is a high value compared to other superconductors such as Ta and Nb, due to the large τ_0 in Al [12].

We have measured the fluctuations in the resonator amplitude in equilibrium at the resonant frequency using a microwave power of -77 dBm. The power spectral density, corrected for system noise, is shown in Fig. 2(a) for various temperatures, which is the central result of this Letter. In Fig. 2(b) the system noise spectrum is shown, which is subtracted from the spectrum measured on resonance, to get the corrected spectra in Fig. 2(a). Parts of the time domain signal where large energy impacts are observed are removed from the analysis as shown in Fig. 2(c), because they distort the dynamic equilibrium. These impacts do not appear in the system noise signal and decay

within a time τ_r, which shows they are events that create quasiparticles, for example, cosmic ray hits [22].

In Fig. 2(a) we observe that the measured power spectral density of the fluctuations has a constant level for all temperatures. The roll-off in the spectra can be described with a single time scale that decreases with temperature. From these two properties we conclude that we directly observe quasiparticle number fluctuations.

The recombination time τ_r is extracted from the measured noise spectra and shown as the black squares in Fig. 3 as a function of temperature. At temperatures from 180–300 mK, we find the expected exponential temperature dependence. Equation (2) is used to fit for the characteristic electron-phonon interaction time τ_0 and we find a value of 458 ± 10 ns, in reasonable agreement with other studies [12,23]. Because of the phonon trapping effect, which we cannot estimate accurately, the measured τ_0 may differ from the pure electron-phonon time [14,23]. At temperatures <160 mK we measure a temperature independent quasiparticle recombination time of 2.2 ms, which is among the longest reported for a thin superconducting film. Alternatively, the recombination time is measured by monitoring the restoration of equilibrium after a short pulse of optical photons [4], which is shown by the filled circles in Fig. 3. The lifetimes obtained from the noise spectra are equal to the lifetimes from the pulse measurement up to 220 mK, indicating that both measurements really probe quasiparticle recombination in equilibrium. The lifetime from the noise spectra agrees well with theory (full line) up to 300 mK. The lifetime from the pulse method shows a deviation from theory, which we always observe in Al on sapphire resonators. The deviation between the two sets of data calls thus for a future analysis of the physical processes in the pulse method.
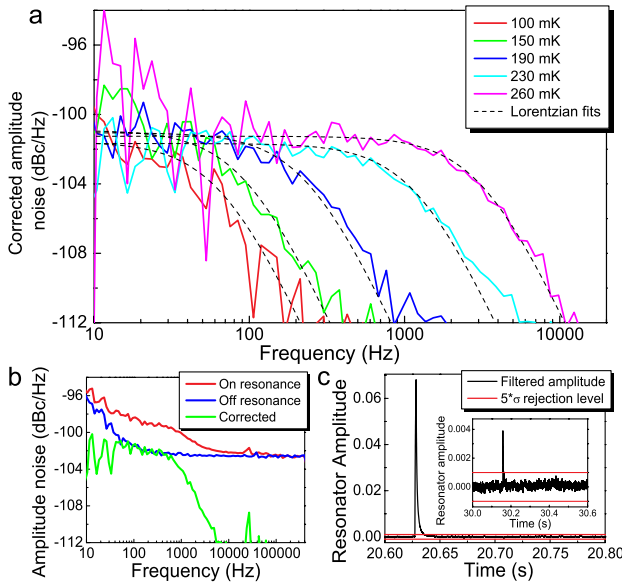

FIG. 2 (color online). (a) Corrected power spectral density of the resonator amplitude fluctuations as a function of frequency, calibrated for system noise contributions as depicted in (b). The spectra are plotted for six different temperatures. Note that the spectral density is temperature independent up to the roll-off frequency. The Lorentzian fits (dashed lines) show that the spectra can be described with a single time scale. (b) The system noise is subtracted from the noise spectrum on resonance. The system noise is obtained by taking a calibration measurement at a frequency far from the resonant frequency as indicated in Fig. 1(c). (c) Part of the time trace of the resonator amplitude, filtered with a moving average filter with a time constant $\tau_r/2$. High energy impacts are observed every 20–30 s. Smaller impacts (inset) happen every 5–10 s. Parts with impacts larger than 5 times the standard deviation (0.43 eV) are rejected.
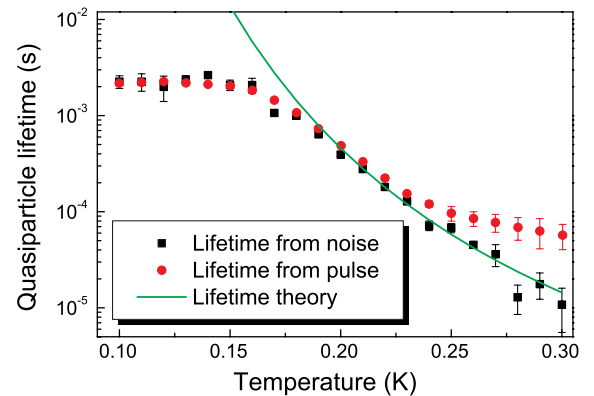

FIG. 3 (color online). The quasiparticle lifetime, obtained from the roll-off frequency of the resonator amplitude noise spectrum (Fig. 2), as a function of temperature. The solid line is the lifetime, calculated from theory. Additionally, the lifetime is determined with a short pulse of light. The exponential decay of the excitation is fit, and the obtained decay time is shown. The measurements show consistently that the lifetime saturates to about 2.2 ms below 160 mK. The error bars represent statistical uncertainties obtained from the fitting procedure.
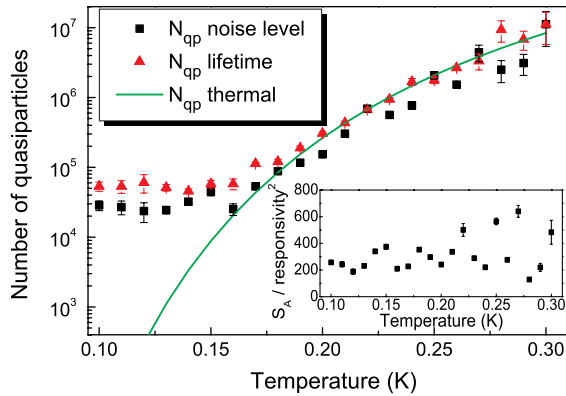
FIG. 4 (color online). The number of quasiparticles as determined from the noise level as a function of temperature. The inset shows that the noise level (S_A) divided by the responsivity is almost temperature independent. The error bars indicate combined statistical uncertainties. Alternatively, the number of quasiparticles in the superconductor is determined from the measured quasiparticle lifetimes. The number of quasiparticles saturates at around 30 000–50 000 below 160 mK, where an exponential decrease is expected (solid line).

We combine the level of the power spectral density (Fig. 2) and the quasiparticle lifetime obtained from the roll-off in this spectral density (Fig. 3) to obtain the number of quasiparticles N_{qp} by using Eqs. (3) and (4) together with the measured values of dA/dN_{qp}. The result is plotted in Fig. 4 with the black squares. As a cross-check, we convert the quasiparticle lifetime measured from the noise roll-off directly into quasiparticle number by using Eq. (2), which is shown in Fig. 4 with red triangles. We assume that the relevant volume is the central strip volume of the resonator. The quasiparticle number, obtained via these two methods, consistently shows a saturation, giving a low temperature quasiparticle density of 25–55 μm^{-3}. We conclude that the quasiparticle lifetime saturation is due to a saturation in the quasiparticle density, consistent with the conjecture of Martinis *et al.* [8].

The question of what the source of the nonthermal [in view of Eq. (1)] quasiparticle density below 160 mK is remains. In literature, it is usually referred to as nonequilibrium quasiparticle density, which is inferred from saturating tunnel rates [6,24] and attributed to electromagnetic noise [25,26] and radiation [27] or, if the first two are eliminated, to cosmic rays, local radioactivity, slow heat release, or stray light [8]. In our experiment, excess quasiparticles due to cosmic ray hits are excluded as shown in Fig. 2(c). Local radioactivity is also excluded as far as it generates similar distinguishable events. The sample box and coaxial cables are thoroughly shielded from stray light, validated by the observation that τ_r is independent of the 4 K-stage temperature, as explained before. Additionally, if there would still be stray light, it would cause an additional photon shot noise [4] contribution, which decreases with increasing temperature, due to the decreasing quasiparticle lifetime. The observed temperature independent noise

level therefore excludes stray light. We cannot completely exclude quasiparticle generation by the microwave signal. The microwave power range over which we can measure the quasiparticle fluctuations is only 4 dB due to limitations in resonator power handling and amplifier noise temperature. Over this range the quasiparticle density is power independent.

For radiation detectors, the noise equivalent power (NEP) is a common way to express the sensitivity. From the noise level and lifetime measurements we determine [3] an electrical NEP of 3.3×10^{-19} WHz$^{-1/2}$ due to generation-recombination noise. For Al resonators with this geometry and the observed remnant quasiparticle density, this is the fundamental limit to the sensitivity. A significant improvement is only possible if one could reduce the remnant quasiparticle density or the resonator volume.

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