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Strong reduction of quasiparticle fluctuations in a superconductor due to decoupling of the quasiparticle number and lifetime

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We measure temperature-dependent quasiparticle fluctuations in a small Al volume, embedded in a NbTiN superconducting microwave resonator. The resonator design allows for readout close to equilibrium. By placing the Al film on a membrane, we enhance the fluctuation level and separate quasiparticle effects from phonon effects. When lowering the temperature, the recombination time saturates and the fluctuation level reduces by a factor ~ 100 . From this we deduce that the number of free quasiparticles is still thermal. Therefore, the theoretical, inverse relation between the quasiparticle number and recombination time is invalid in this experiment. This is consistent with quasiparticle trapping, where on-trap recombination limits the observed quasiparticle lifetime.

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In a superconductor well below its critical temperature, most electrons are bound together in Cooper pairs, while the number of unpaired electrons, quasiparticles, exponentially decreases with decreasing temperature. The recombination time of the quasiparticles is inversely proportional to the quasiparticle number and therefore increases exponentially towards lower temperature. At a constant, finite temperature, the quasiparticle number fluctuates around its average value due to random generation and recombination events [1]. In Ref. [2], a measurement of these fluctuations showed that the quasiparticle number and recombination time are indeed inversely proportional, and as a consequence the quasiparticle fluctuation level is *constant* as a function of temperature. Both observations are consistent with direct recombination of free quasiparticles with the emission of a phonon [3] [see Fig. 1(a)]. Also in the presence of excess quasiparticles at low temperature, which often occur in superconducting devices in varying conditions [4–9], the relation between the number of quasiparticles and their recombination time is maintained [2].

In this Letter we show experimentally that the intimate relation between the free quasiparticle number and recombination time is broken when quasiparticles are first trapped and then recombine as depicted in Fig. 1(b).

Impurities and disorder in superconductors are known to reduce the recombination time at low temperatures [10,11] and can fundamentally change the relation between the quasiparticle number and recombination time. The low-temperature recombination time is typically limited from tens of microseconds [11,12] to milliseconds [2,13], depending on the material. However, these experiments only measure the recombination time from a nonequilibrium pulse decay, or suffer from excess quasiparticles. Moreover, in tunnel junc-

tion devices, including qubits, only nonequilibrium, local quasiparticle properties can be measured directly. Understanding the reduced recombination time at low temperatures and close to thermal equilibrium, and in particular its relation to the quasiparticle density, has been hindered by the lack of a sensitive probe of quasiparticle dynamics.

Here, we measure quasiparticle fluctuations in a small Al volume ($27 \mu\text{m}^3$), embedded in a NbTiN superconducting resonator. This design allows for readout at low microwave powers, minimizing the creation of excess quasiparticles [14]. By placing the Al volume on a membrane, we enhance the fluctuation level and separate quasiparticle effects from phonon effects.

We observe a strong reduction in the quasiparticle fluctuation level by a factor ~ 100 when lowering the bath temperature from 350 to 200 mK, together with a saturation of the recombination time. Hence, the inverse proportionality between quasiparticle number and recombination time must be broken and the recombination time is not limited by excess quasiparticles as we observed before [2,14]. Together, our

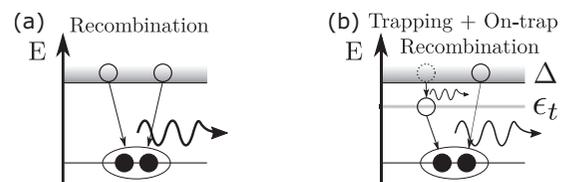


FIG. 1. Schematic representations of simple recombination (a) and trapping with subsequent on-trap recombination (b) of two quasiparticles. Δ is the superconducting energy gap and ϵ_t the trap energy. Curved arrows represent emitted phonons, with thickness indicating their relative energy.

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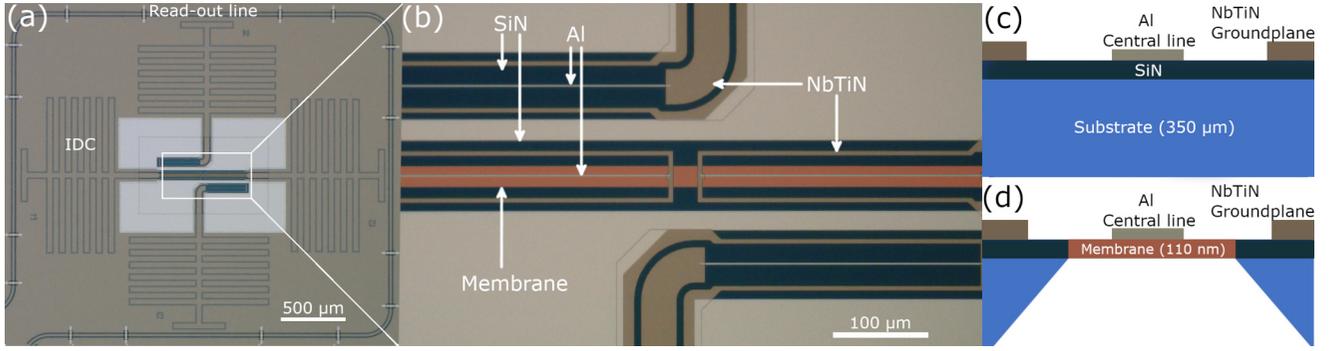


FIG. 2. (a) Micrograph of the resonators studied. (b) Zoom-in on the Al inductive parts of the resonators, of which two are on a 110-nm-thick SiN membrane (highlighted in orange). (c), (d) Schematic cross sections (not to scale) of the inductive (Al) part of a substrate and membrane resonator, respectively. The coplanar waveguide (CPW) dimensions are such that only the Al central line is suspended by the membrane.

observations are in qualitative agreement with quasiparticle trapping and subsequent on-trap recombination.

Our methodology is well suited to better understand the quasiparticle dynamics in devices where quasiparticle trap structures [15,16] and vortices [17] are introduced deliberately, to reduce the excess quasiparticle density in critical regions of a device.

In thermal equilibrium and at low temperatures ($T \ll \Delta/k_B$), the number of quasiparticles in a superconducting volume V is given by [18]

$$N_{\text{qp}}(T) = 2VN_0\sqrt{2\pi k_B T \Delta} e^{-\Delta/k_B T}, \quad (1)$$

where N_0 is the single spin density of states at the Fermi level (we use $N_0 = 1.72 \times 10^4 \mu\text{eV}^{-1} \mu\text{m}^{-3}$, for Al), k_B is the Boltzmann constant, and $\Delta = 1.76k_B T_c$ is the superconducting gap energy, with T_c the critical temperature. The intrinsic quasiparticle lifetime with respect to recombination (hereafter called “the quasiparticle lifetime”) is given by [3]

$$\tau_{\text{qp}}(T) = \frac{\tau_0 N_0 V (k_B T_c)^3}{2\Delta^2 N_{\text{qp}}(T)} = \frac{V}{RN_{\text{qp}}(T)}, \quad (2)$$

where τ_0 is a material-dependent characteristic time for the electron-phonon coupling. For Al, we take $\tau_0 = 0.44 \mu\text{s}$ [3]. In the last equality, all the material-dependent parameters are combined into the recombination constant R .

In experiments, the relaxation of an ensemble of quasiparticles is typically probed. As the recombination of two quasiparticles into a Cooper pair is a pairwise process and the emitted phonon can subsequently break a Cooper pair [19], the *apparent* quasiparticle lifetime is given by

$$\tau_{\text{qp}}^* = \tau_{\text{qp}}(1 + \tau_{\text{esc}}/\tau_{\text{pb}})/2. \quad (3)$$

Here, τ_{esc} is the phonon escape time, τ_{pb} is the phonon pair-breaking time, and the factor in parentheses is called the *phonon trapping factor*. Equation 3 is valid when $\tau_{\text{esc}}, \tau_{\text{pb}} \ll \tau_{\text{qp}}$, which is typically the case [20]. We take $\tau_{\text{pb}} = 0.28 \text{ ns}$ for Al [3]. τ_{esc} can experimentally be tuned [21,22], for instance, with the use of a membrane, which we use here to distinguish phonon effects from intrinsic quasiparticle processes.

Fluctuations in the quasiparticle number occur randomly. Starting from a master equation approach [1], the power spectral density (PSD) of these fluctuations can be calculated

to be

$$S_{N_{\text{qp}}}(\omega) = \frac{4\tau_{\text{qp}}^* N_{\text{qp}}}{1 + (\omega\tau_{\text{qp}}^*)^2}. \quad (4)$$

This is a Lorentzian spectrum, with a constant level with temperature (since $\tau_{\text{qp}}^* \propto 1/N_{\text{qp}}$) and a roll-off frequency of $\omega = 1/\tau_{\text{qp}}^*$.

The device under study is shown in Fig. 2. Four NbTiN-Al hybrid [23] resonators are capacitively coupled to a readout line patterned around them. The capacitive part is a NbTiN double-sided interdigitated capacitor (IDC) [24,25] and the inductive part is a NbTiN coplanar waveguide with an Al central line. For details, see the Supplemental Material [26].

The resonator response is only sensitive to quasiparticle–Cooper-pair fluctuations within the Al ($V = 27 \mu\text{m}^3$ and $T_c = 1.26 \text{ K}$), as we measure at $T \ll T_{c,\text{NbTiN}} = 15.6 \text{ K}$ and the current density is much higher in the Al section due to the IDC design. Furthermore, as $\Delta_{\text{Al}} \ll \Delta_{\text{NbTiN}}$, quasiparticles are confined to the Al volume.

The resonators are mounted in a pulse tube precooled adiabatic demagnetization refrigerator, surrounded by a CRYOPHY and a superconducting magnetic shield, and isolated from stray light by a “box-in-a-box” configuration [27]. The forward microwave transmission S_{21} is recorded at an on-chip read power, $P_{\text{read}} = -99 \text{ dBm}$. In the following, we present the results for two resonators, of which one has its sensitive volume on a membrane and one on the substrate [see Figs. 2(c) and 2(d)].

We translate the complex S_{21} to an amplitude (A) and phase (θ) to distinguish changes in dissipation (A) and kinetic inductance (θ). These variables are defined relative to the resonance circle, which is measured for each temperature before the fluctuation measurement. The responsivity is given by $dX/dN_{\text{qp}} = 2\alpha_k Q\kappa_X/V$, where X is either A or θ and α_k is the fraction of kinetic inductance over the total inductance. κ_X describes the change in complex conductivity with respect to a change in quasiparticle density, which is only weakly dependent on temperature [28]. Q is the loaded quality factor and measured to be 4.3×10^4 and 3.5×10^4 at 50 mK, for the membrane and substrate resonator, respectively. The resonators are designed to have the same volume and kinetic inductance fraction.

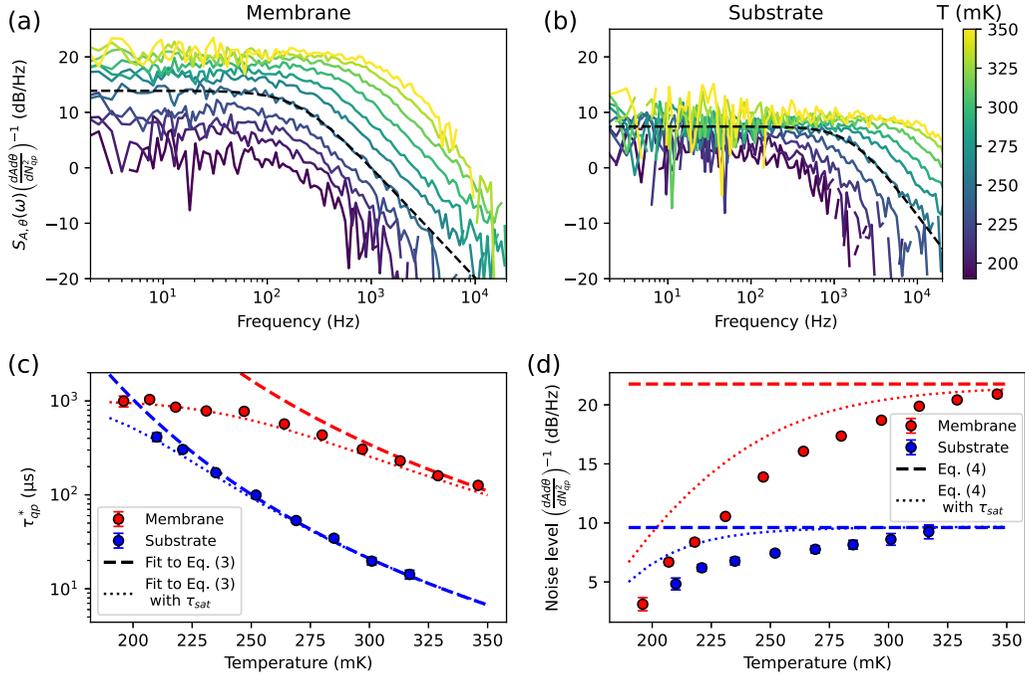


FIG. 3. (a), (b) Measured quasiparticle fluctuations for a resonator (a) on the membrane and (b) on the substrate. The quasiparticle fluctuations are determined from the measured cross-PSD, using the measured responsivities and Eq. (5). The measurements are performed at -99 dBm readout power. The dashed black lines give examples of the Lorentzian fits [Eq. (4)]. (c) Lifetimes and (d) noise levels from Lorentzian fits [Eq. (4)] to the spectra in (a) and (b). The error bars indicate statistical uncertainties from the fitting procedure. Only fits with a relative fitting uncertainty lower than 13.5% in lifetime are displayed. The dashed lines in the lifetime plot are fits to Eqs. (2) and (3) for temperatures ≥ 300 mK, with τ_{esc} as the free parameter. The dashed lines in the level plot are calculated from Eq. (4), with Eqs. (1) and (3) and the same τ_{esc} . The dotted lines are calculated in the same way, but with $\tau_{qp}^* \rightarrow [(\tau_{qp}^*)^{-1} + (\tau_{sat})^{-1}]^{-1}$, with $\tau_{sat} = 1$ ms.

We measure 40-s time streams at 50 kHz of A and θ , at temperatures ranging from 50 to 400 mK. We filter pulses caused by cosmic rays [7] or other external sources [26] and calculate the cross-PSD, $S_{A,\theta}(\omega)$. By using the cross-PSD, we extract the dissipation-kinetic inductance (i.e., quasiparticle–Cooper-pair) fluctuations and suppress uncorrelated noise sources such as amplifier and two-level system (TLS) noise [14]. We determine the spectrum of the quasiparticle fluctuations via

$$S_{N_{qp}}(\omega) = S_{A,\theta}(1 + (\omega\tau_{res})^2) \left(\frac{dAd\theta}{dN_{qp}}\right)^{-1}, \quad (5)$$

where $\tau_{res} = Q/\pi f_0$ is the resonator ring time (f_0 is the resonance frequency), typically a few μ s. This implies $(\omega\tau_{res})^2 \ll 1$ for frequencies below 100 kHz and we therefore neglect this factor. The last factor in Eq. (5) is the multiplication of the amplitude and phase responsivities, which we determine from a measurement of $S_{21}(\omega; T)$, for $T > 250$ mK [26].

The central result of this Letter is presented in Fig. 3. Figures 3(a) and 3(b) show the measured cross-PSDs and Figs. 3(c) and 3(d) show the extracted apparent quasiparticle lifetime and fluctuation level from Lorentzian fits to these spectra [Eq. (4)]. For high temperatures (≥ 300 mK), we observe a higher level and τ_{qp}^* for the membrane resonator compared to the substrate resonator. This is expected from Eqs. (3) and (4), as the membrane effectively increases τ_{esc} . The dashed lines in Figs. 3(c) and 3(d) are fits to Eqs. (2) and (3) for temperatures ≥ 300 mK, with τ_{esc} as only free parameter, resulting in $\tau_{esc} = 5.6 \pm 0.4$ ns and 0.09 ± 0.02 ns

for the membrane and substrate resonator, respectively. These intervals only indicate uncertainties from the fitting procedure. Equivalently, the phonon trapping factors [Eq. (3)] are 21 and 1.3 from which we would expect a 12 dB higher fluctuation level for the membrane resonator. This is indeed observed in Fig. 3(d). The dashed lines give the expected level from Eq. (4), where the fitted τ_{esc} is used. A detailed analysis of the effects of the membrane on phonon statistics and energy resolution are published elsewhere [29].

The fluctuation levels expected from Eq. (4) [dashed lines in Fig. 3(d)] are constant with changing temperature. In sharp contrast, we here observe a strongly decreasing level when lowering the temperature. At temperatures < 190 mK, no Lorentzian spectrum [Eq. (4)] can be identified. The individual amplitude and phase PSDs show this behavior as well [26]. As the responsivity factor in Eq. (5) is constant in this temperature range [26], this result should be interpreted as a strong reduction of $S_{N_{qp}}$. We calculate N_{qp} from $S_{N_{qp}}$ by dividing the level by $4\tau_{qp}^*$, which is shown in Fig. 4. The number of quasiparticles follows a thermal dependence for both the substrate and membrane resonator. Therefore, we conclude that the lifetime saturation is not caused by excess quasiparticles, in contrast to the observations in Ref. [2]. On top of that, the lifetimes observed for membrane and substrate resonators saturate at a similar level despite the 16 times stronger phonon trapping in the membrane. Therefore the lifetime saturation must originate from an effect that directly interacts with the quasiparticle system.

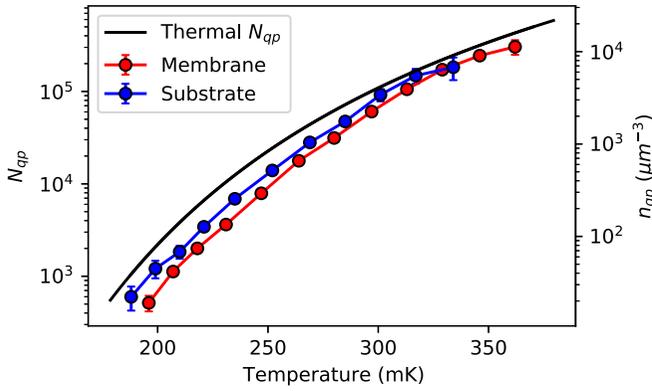


FIG. 4. Number of quasiparticles and quasiparticle density (right axis) within the Al volume, calculated from the Lorentzian fits in Figs. 3(c) and 3(d). Error bars indicate propagated fitting uncertainties. The solid black line is from Eq. (1).

The design of the NbTiN-Al hybrid resonators enables readout low microwave powers (P_{read}), which minimizes the creation of excess quasiparticles. When we increase P_{read} , we eventually observe excess quasiparticles without a level reduction, equivalent to a higher, P_{read} -induced, effective temperature. The results of Refs. [2,14] are thus recovered in the high P_{read} regime. See the Supplemental Material for more details [26].

To verify that the lifetime saturation without the creation of excess quasiparticles results in a level reduction, we calculated the lifetimes and fluctuation levels with Eq. (4), but with an alteration to Eq. (3),

$$\frac{1}{\tau_{\text{qp}}^*} = \frac{2}{\tau_{\text{qp}}(1 + \tau_{\text{esc}}/\tau_{\text{pb}})} + \frac{1}{\tau_{\text{sat}}}, \quad (6)$$

with τ_{sat} the saturation lifetime. For both the membrane and substrate, τ_{sat} is set to 1 ms, based on τ_{qp}^* from the membrane resonator. The results are plotted in Figs. 3(c) and 3(d) as the dotted lines, which follow the measured τ_{qp}^* very well and shows a decrease in fluctuation level for both resonators. The deviations of level in Fig. 3(d) are likely caused by the responsivity measurement method [26].

A possible cause of a lifetime saturation without excess quasiparticles is *quasiparticle trapping*. From theory, it is known that magnetic [30–33] and nonmagnetic [34–36] impurities can cause subgap electronic states [37] and disorder, which can result in local variations of Δ [38,39]. Also, thickness variations [40] and unpaired surface spins [41] due to native oxide [42] may induce Δ variations. Quasiparticles can be trapped at these suppressed Δ regions [43,44] by inelastic scattering, resulting in a background number of trapped quasiparticles N_t . At low temperatures, N_{qp} [Eq. (1)] inevitably becomes comparable to N_t , leading to dominating quasiparticle trapping behavior even for low impurity concentrations, weak disorder, or other effects that lead to small variations in Δ [43,45,46].

At that point, the quasiparticle lifetime is no longer limited by free quasiparticle recombination, but by *on-trap* recombination events, where a free quasiparticle recombines with

a trapped one. Trapping itself does not change the number of Cooper pairs and therefore is not observable in our experiments. This is in contrast to experiments where the free quasiparticle density near a junction is measured, in which case trapping dominates the low-temperature behavior [47,48]. The trapping states we conjecture are dissipative, as the amplitude and phase PSDs show the same temperature behavior [26], in contrast to what is observed for disordered TiN [11,49].

Analogous to Eqs. (3) and (2), the saturation lifetime due to on-trap recombination can be written $\tau_{\text{sat}} = V/2R_t N_t$, with R_t the on-trap recombination constant [45]. The phonon that is emitted during an on-trap recombination event has an energy $\Omega < 2\Delta$ and is therefore unable to break a Cooper pair into two free quasiparticles. *On-trap* pair breaking (a subgap phonon breaking a Cooper pair into a trapped and free quasiparticle) is far less likely because the density of trapping states is assumed to be small [45]. Therefore, τ_{sat} is independent of phonon trapping, which is consistent with the observation that the substrate and membrane resonator show the same τ_{sat} .

To investigate where quasiparticle traps could be located in our system, we conducted the same experiment with different geometries of the Al strip. Width (0.6–1.5 μm) and length (0.12–1.4 mm) variations did not affect the saturation lifetime, which implies the traps are not located at the NbTiN-Al interface or the sides of the Al central line. τ_{sat} increases from 0.7 to 3.1 ms with increasing the film thickness from 25 to 150 nm, respectively, and the reduction in fluctuation level is not observed for the thickest film. The experimental data of these geometry variations are presented in the Supplemental Material [26]. This suggests that the trap density (N_t/V) decreases for thicker films and the location of the traps is either at the Al-substrate interface or the top surface. The top surface might contain unpaired surface spins from the native Al oxide [10,42]. Further progress in understanding the trapping mechanism will require identification of the microscopic origin of the traps.

We here analyzed the lifetime saturation phenomenologically using Eq. (6). A more detailed model of the fluctuations comprises multivariable rate equations for quasiparticles and phonons, including quasiparticle trapping and on-trap recombination terms [1,45]. Only nonequilibrium experiments [17,48,50] were conducted previously ($\delta N_{\text{qp}} \gg N_{\text{qp}}$), which allows one to neglect the generation terms and feedback from the phonon system, in contrast to our near-equilibrium ($\delta N_{\text{qp}} \ll N_{\text{qp}}$) experiment. We have studied several such models [51], but without satisfactory results due to the number of (unknown) input parameters. However, we can constrain such a model to follow our experimental data.

Quasiparticle trapping could decrease the low-temperature responsivity of superconducting tunnel junction detectors [47], cause long-lived excitations in granular Al resonators [50], and lead to an anomalous electrodynamic response of disordered TiN resonators [11,49]. However, for single-photon detection using microwave kinetic inductance detectors (MKIDs) [52,53], the quasiparticle trapping effects can be beneficial. The fluctuation level reduction results in a higher signal-to-noise ratio when generation-recombination noise is the dominant noise source and the responsivity

stays the same, as we observe [26,29]. This means, somewhat counterintuitively, that single-photon detector performance can be improved at low temperatures, by introducing quasiparticle traps, compared to a quasiparticle number saturation [14].

In conclusion, at high temperatures (>300 mK), quasiparticle lifetimes in Al superconducting resonators are well described by a simple recombination in thermal equilibrium [3]. At low temperatures, the lifetime is limited to 1 ms, independent of phonon trapping, while the quasiparticle number exponentially decreases with temperature. This results in a strong reduction of quasiparticle fluctuations. Quasiparticle trapping with subsequent on-trap recombination is consistent with our observations. Varia-

tions in the Al strip geometry (width, length, and thickness) showed that the traps are likely located at top or bottom surfaces.

All presented data and used analysis scripts are openly available [62].

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