Coherence Time Enhancement of Interacting Two-Level Systems in Aluminum Superconducting Resonators

Tamin Tai,1,* Jingnan Cai,1 and Steven M. Anlage1

1Quantum Materials Center, Department of Physics, University of Maryland, College Park, MD 20742-4111, USA

(Dated: September 24, 2021)

Superconducting resonators are widely used in many applications such as qubit readout for quantum computing applications, and kinetic inductance detectors. These resonators are susceptible to numerous loss and noise mechanisms under microwave excitation, especially the dissipation due to non-equilibrium quasi-particles and two-level systems (TLS), which can result in a decrease of the superconducting intrinsic quality factor ($Q_i$) in high quality superconducting resonators. Particularly in the few-photon and low temperature ($T$) regime, TLS losses can become a dominant loss mechanism. In this study, novel aluminum half-wavelength resonators are investigated, focusing on the loss properties at extra-low power and low temperature. An unusual increase of $Q_i(T)$ with deceasing temperature is observed. This behavior is attributed to the increase of TLS coherence time ($T_2$) at ultra-low temperatures and powers. This $T_2$ increase is consistent with other work on resonant frequency noise in resonators and measurements of individual TLS, and likely arises from interacting TLS in the aluminum half-wavelength resonators.

I. INTRODUCTION

Two-dimensional (2D) planar high internal quality factor ($Q_i$) superconducting resonators have been widely fabricated and investigated in recent times for such application as single photon detectors,[1] kinetic inductance detectors,[2] and quantum computing technology.[3] Recently, these high $Q_i$ resonators have been widely applied in the design of large scale quantum gates, and tremendous progress has been made in terms of design, fabrication and measurement techniques, which has led to orders of magnitude increase in coherence time and improved quantum fidelity [3–5]. In microwave measurements, although all qubits are operated at an excitation frequency well below the superconducting gap energy, microwave photons can be absorbed by quasiparticles, and these quasiparticles interact with a phonon bath, creating non-equilibrium distributions of both quasiparticles and phonons. As a result, Cooper pair-breaking phonons can be created.[6–9] This process contributes non-equilibrium quasiparticles, in addition to pair-breaking photons induced by cosmic rays,[10] higher order microwave harmonics, and the possibility of stray infrared radiation [11–13]. These non-equilibrium quasiparticles are a limiting factor on superconducting resonator $Q_i$ and qubit coherence under any level of microwave excitation, and their presence results in a decrease of qubit relaxation time ($T_1^{Qubit}$) and coherence time ($T_2^{Qubit}$).[14]

Moreover, comparable losses due to two-level systems (TLS) almost universally occur [15–22] in the low power regime in 2D superconducting resonators. It is widely believed that TLS are due to a fluctuating electric dipole that couples to microwave electric fields. In general, TLS could originate from three kinds of interfaces on the devices. One is the metal-vacuum interface due to surface oxide or contaminants. The other is the metal-dielectric substrate interface due to residual resist chemicals, and buried adsorbates. The third is the dielectric substrate-vacuum interface which could have hydroxide dangling bonds, processing residuals, and adsorbates.[23] To address these issues, different kinds of geometry of coplanar waveguide (CPW) structure have been proposed and fabricated in the design of these resonant devices, with more care given to the surface treatment to alleviate the TLS losses [24]. For example, a trenched structure in the CPW helps to mitigate the metal-dielectric TLS interaction with the resonator fields.[25] These efforts have improved the 2D resonator intrinsic quality factor to more than 1 million in recent realizations of high-$Q_i$ resonators [25–28]. Nevertheless, TLS still exist even in extra high $Q_i$, 3D superconducting radio frequency cavities used in particle accelerator applications.[29] Recently other sources of TLS loss have been proposed based on quasiparticles trapped near the surface of a superconductor.[30]

Clearly TLS-based loss is a universal issue in superconducting resonators. The behavior of TLS under extra-low microwave power and low temperature still lacks full elaboration due to the constraints of noise floor levels in electronic equipment. Therefore, understanding this TLS behavior especially in the low microwave excitation regime is essential, and would assist the superconducting quantum information community to mitigate its effect on operating quantum devices.

We have designed a 2D half wavelength resonator with a tapering geometry for the center line in a CPW structure, where the linewidth varies from 50 µm down to 1 µm. The reason for this design is to host many three-junction flux qubits in the center of the transmission line resonator. This design can provide strong radio frequency coupling to the qubits to allow study of the collective behavior of quantum meta-materials. In this way,
qubits serve as artificial meta-atoms and are analogous to atomic cavity quantum electrodynamics, and coupling between neighboring artificial meta-atoms [31–35] also can be read out through the dispersive frequency shift of the cavity. [36–38] Theoretical publications discussing the physics of qubit arrays coupled to the harmonic cavities predict a number of novel collective behaviors of these meta-atoms [39–41]. In this paper, we report our novel finding on the TLS behavior in ultra-low power and low temperature through the design, fabrication, and characterization of this special geometry half-wavelength resonator, without the qubits. The technique of ultra-low power microwave measurement with low noise to enhance the signal-to-noise ratio (SNR) is critical for measuring this TLS behavior.

II. EXPERIMENTAL METHODS

An aluminum (Al) half-wavelength (λ/2) CPW resonator on sapphire substrate was designed with a center line width \( w = 50 \, \mu m \) and spacing \( s = 30 \, \mu m \) (the distance between center conductor line and ground plane as illustrated in Fig. 1(b) to maintain the characteristic impedance near 50 Ω in the meander part. At the center of the resonator a tapering structure narrows the center line width down to \( w = 1 \, \mu m \) and spacing to \( s = 12 \, \mu m \), which gradually increases the characteristic impedance to 100 Ω at the resonator center. Fig. 1 (a) shows a perspective view of the resonator in a diced chip with a designed fundamental frequency around 3.6 GHz. The entire resonator is surrounded by many 10 μm by 10 μm vortex moats. The resonator is symmetric and capacitively coupled through 5 μm capacitive gaps (1 (b) and 1 (c)) in the center conductor. A topographic image of the narrowed resonator center section is shown in Fig. 1 (d) with a critical dimension around \( w = 1 \, \mu m \) in width.

This CPW resonator was fabricated using standard photo-lithography technology procedures. In detail, first a 100 nm thick Al film was deposited on a 3-inch diameter sapphire wafer using thermal evaporation technology with a background pressure of \(~ 3 \times 10^{-7} \) mbar. Then a thin SHIPLEY1813 photo-resist was coated on top of the film and exposed to UV through the designed photomask. The UV exposed wafer was developed and then wet etched by commercial Transene Aluminum Etchant.
The remaining photoresist was stripped off by acetone and the entire wafer was cleaned by methanol and isopropanol. Finally, the wafer was coated in a protective photo-resist and then diced into many chips. After dicing, the protective photo-resist was removed and the chip was mounted on a printed circuit board bolted inside a copper box. Several lumps of indium were pressed between the on-chip ground planes and the copper box ground to achieve a continuous ground contact, which mitigates parasitic resonant microwave modes due to uneven electrical grounding. The indium lumps also secured the chip in the center of the printed circuit board. The on-chip transmission line is wire-bonded to the center conductor of the transmission line on the printed circuit board by gold wires. Finally, the copper box is capped by a copper lid to eliminate stray light illumination.

The device was placed in a closed Cryoperm cylinder in a BlueFors (BF-XLD 400) cryogen-free dilution refrigerator (base temperature 10 mK) to minimize any stray DC magnetic field, and the shield was thermally anchored to the mixing chamber plate. The microwave excitation was attenuated by 66 dB in the input line through a cryogenic amplifier with 36 dB gain and then boosted by another 37 dB at ambient temperature before the transmitted signal \( S_{21} \) was measured by a Keysight N5242A vector network analyzer (VNA). The ultra-low power measurements were performed using the smallest intermediate frequency bandwidth (1 Hz) of the VNA, with a 400 kHz span across the resonance, following 5 averages to reduce the random noise. Further details of the experimental setup for the high SNR measurement at ultra-low microwave power can be found in section I of the Supplemental Material.

III. EXPERIMENTAL DATA

The measured transmitted signal \( S_{21}(f) \) has a fundamental \((\lambda/2)\) resonance peak around \( f = 3.644 \text{ GHz} \) at the fridge base temperature when sweeping the frequency, \( f \), (or \( \omega = 2\pi f \)). The complex \( S_{21}(f) \) signal is fit to an equivalent circuit model of a two-port resonator capacitively coupled to external microwave excitation [9, 42].

\[
S_{21}(f) = |S_{21,in}| |S_{21,\text{out}}| \left( \frac{Q_L/Q_c}{1 + 2jQ_L(f_0 - 1)} e^{j\phi} \right) + C_0 \tag{1}
\]

where \( |S_{21,in}| \) is the attenuation loss in the transmission of the input line, \( |S_{21,\text{out}}| \) is the gain in the transmission of the output line, \( Q_L \) is the loaded quality factor, \( Q_c \) is the coupling quality factor representing the dissipation to the external circuit, \( j = \sqrt{-1}, f_0 \) is the resonance frequency of the half-wavelength \((\lambda/2)\) CPW resonator, \( \phi \) is the phase and \( C_0 \) is an offset in the complex \( S_{21} \) plane due to background contributions.[42] The internal quality factor, \( Q_i \), representing internal dissipation is extracted from the identity \( 1/Q_L = 1/Q_i + 1/Q_c \). The average number of circulating microwave photons in the cavity can be estimated using the approximation [9, 43] \( < n > = \frac{2Q_i^2 P_{in}}{Q_e \omega_0} \) in two ports measurement where \( P_{in} \) is the input power to the resonator after the attenuation in the input line, \( h \) is the reduced Planck constant and \( \omega_0 = 2\pi f_0 \) is the angular frequency of the resonance.

Figure 2(a) shows the temperature dependence of the resonant frequency, \( f_0 \), for different circulating microwave photon numbers inside the CPW resonator, by fitting each resonance circle in the complex \( S_{21} \) plane to Eq. 1. For each case, the maximum resonance frequency occurs at the 10 mK fridge base temperature and then shows a local maximum around 200 mK. This phe-
nomenon seems to be independent of the average circulating photon number. When the fridge temperature is higher than 200 mK, the resonance frequency quickly decreases with increasing temperature due to the large injection of thermal quasiparticles, which results in a surface impedance increase in the superconducting resonator. The dependence of internal quality factor $Q_i$ on the average circulating photon number at different representative fridge temperatures is shown in Fig. 2 (b). For each curve, the $Q_i$ gradually increases with increased circulating photon numbers, but then the $Q_i$ is suddenly suppressed at extremely high photon numbers ($< n > \sim 10^8$). One important observation is that in the low photon number regime, one can see the $Q_i$ at $T = 12$ mK is higher than the $Q_i$ at $T = 80$ mK and $T = 120$ mK. This unusual behavior will be discussed in the next section.

IV. MODELING

Researchers working on the loss mechanisms of superconducting resonators have primarily attributed the power and temperature dependent frequency shifts and $Q_i$ variations at low temperatures to the non-equilibrium quasiparticles in the superconducting metals and TLS in the dielectrics. However, it can be difficult to experimentally distinguish the TLS-loss mechanism from non-equilibrium quasiparticle-related loss, since both loss mechanisms are power and temperature dependent in the same operational regime for many superconducting devices, including resonators and qubits. Here, we will interpret our data in terms of two main features, namely the electrodynamics of TLS in the dielectrics combined with that of both equilibrium and non-equilibrium quasiparticles in the metals. To interpret the local maximum of resonance frequency at 200 mK in Fig. 2, an analytical equation combining the TLS model and thermal quasiparticle model is used to fit the curve.\cite{19, 24, 44}

$$\frac{f(T) - f_n}{f_n} = F \frac{\tan \delta}{\pi} \left( \text{Re} \left[ \Psi \left( \frac{1}{2} + \frac{\hbar \omega}{2 \pi j k_B T} \right) \right] - \log \left( \frac{\hbar \omega}{2 \pi k_B T} \right) \right) - \frac{\alpha}{2} \sqrt{\frac{\pi \Delta_{S0}}{2 k_B T} \exp \left(-\frac{\Delta_{S0}}{k_B T} \right)}$$

where $F$ is the filling factor defined as the fraction of the resonator total electrical energy stored in the TLS material,\cite{17} $f_n$ is the resonance frequency in the limit of low temperature and microwave power, $\Psi(\cdot)$ is the complex digamma function, $\tan \delta$ is the loss tangent of the dielectric material, $k_B$ is the Boltzmann constant, $T$ is temperature, $\alpha = \frac{L_{\text{kinetic}}}{L_{\text{total}}}$ is the kinetic inductance fraction of the CPW resonator, and $\Delta_{S0}$ is the aluminum superconducting gap at zero temperature. The first term in Eq. 2 represents the frequency shift created by the TLS mechanism\cite{17} and the second term is the frequency shift due to equilibrium quasiparticles using the Bardeen-Cooper-Schrieffer (BCS) model\cite{44} [45]. (We show in the Sup. Mat. section III that the non-equilibrium depairing is temperature independent in this range, hence we can ignore this contribution.)

The fit to the frequency shift data is shown in Fig. 3, and the extracted fitting parameters indicate that the aluminum superconducting gap at zero temperature is $\Delta_{S0} = 174.03$ µeV, a value close to the BCS gap approximation which is $1.76 k_B T_c$ with transition temperature $T_c = 1.15$ K. The values of the other fitting parameters are $\alpha = 0.0185$, $f_n = 3.6422 \times 10^9$ Hz and $F \times \tan \delta = 6.4 \times 10^{-6}$. The value of $F \times \tan \delta$ is consistent with other results on a variety of superconducting resonators.\cite{17, 21}. Note that this fit predicts the maxi-

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig3.png}
\caption{Temperature dependent fundamental ($\lambda/2$) mode resonant frequency $f_0(T)$ of the Al CPW resonator at an external microwave excitation creating $\sim 10$ circulating photons. The inset highlights the low temperature regime which the frequency shift is dominated by the TLS mechanism. The dots are experimental data and solid line is the model fit.}
\end{figure}
unusual as not statistically significant. However, this is consistent with previous observations that attribute the difference between predicted and measured $Q$-factors to the difference in bandwidth of TLS states contributing to $Q$ and to frequency shift.[47, 48] Note that similar frequency shift behavior at low temperatures above $200 \text{ mK}$ is also observed in our aluminum resonators at low power and low temperature, an extended TLS model is applied here.

A general TLS Hamiltonian can be written as $H_{\text{TLS}} = \frac{1}{2} \left( -\Delta \, \Delta_0 \right)$ where $\Delta$ is the asymmetry of the double well potential and $\Delta_0$ is the tunneling barrier energy between two potential wells.[9, 15] In addition, one should also expect there are many TLSs with different values of $\Delta$ and $\Delta_0$, therefore in the standard model of TLS loss, a uniform distribution in $\Delta$ and log uniform distribution in $\Delta_0$ is assumed.[15] When the two-level population is in thermal equilibrium, for which the phonon emission and electromagnetic absorption processes are balanced, the longitudinal relaxation time ($T_1$) of the assembled TLS is given by [49]

$$\frac{1}{T_1} = \left( \Delta_0 - \Delta \right)^2 \left[ \frac{\gamma_L^2}{v_L^5} + \frac{\gamma_T^2}{v_T^5} \right] \frac{\varepsilon^3}{2\pi \rho v_T^4} \coth \left( \frac{\varepsilon}{2k_B T} \right)$$

(3)

where $\varepsilon = \sqrt{\Delta^2 + \Delta_0^2}$ is the difference of the two eigenvalues of $H_{\text{TLS}}$, $\gamma_L$ and $\gamma_T$ are the longitudinal and transverse deformation potentials, respectively, $v_L$ and $v_T$ are the longitudinal and transverse sound velocities, respectively, and $\rho$ is the mass density. In addition, the coherence time ($T_2$) of the interacting TLS [50] is described as [15, 49]

$$\frac{1}{T_2} \sim C \frac{2\gamma^2 P_0 k_B T \Delta}{\pi \hbar^2 \varepsilon}$$

(4)

Note that $C$ is dimensionless phonon scattering rate,[51] and is often taken to be a constant of order unity, $P_0 k_B T$ is the volume density of thermally excited TLS with $P_0$ (two-level density of states) on the order of $10^{44} \text{ J}^{-1} \times 10^{}$.
due to quasiparticles with density $n_{neq}$ and electric field independent terms. From this result, one can expect $T_2$ has a $1/T$ dependence on temperature, which arises from the TLS interaction bandwidth scaling with thermal energy $k_B T$.\cite{15}

The imaginary part of the relative dielectric constant is regarded as the TLS loss tangent $n_{TLS}$

$$\delta_{TLS} = \epsilon''_{TLS}(\omega)/\epsilon_{TLS} = \frac{\delta_{TLS}^0 \tanh\left(\frac{\varepsilon}{2k_B T}\right)}{1 + 2\varepsilon T_2} \tag{5}$$

with Rabi frequency $\Omega$\cite{49}

$$\Omega = \frac{2d_0 \Delta_0}{\sqrt{3\hbar \varepsilon}} |\bar{E}| \tag{6}$$

where $d_0$ is the maximum transition electric dipole moment of a TLS and $\bar{E}$ is the microwave electric field. By expressing $T_1, T_2$ and $\Omega$ with Eqs. (3), (4), and (6), the quality factor due to the TLS loss can be written as,

$$\frac{1}{Q_{TLS}} = \frac{1}{Q_{TLS}^0} \tanh\left(\frac{\varepsilon}{2k_B T}\right) \sqrt{1 + 2\tanh\left(\frac{\varepsilon}{2k_B T}\right)}$$

with $\bar{A} = \left(\frac{4d_0^2}{3\varepsilon^2} \left[\frac{\gamma^2 T_1}{v_L^2} + \frac{\gamma^2 T_2}{v_T^2}\right]^{-1} \frac{\varepsilon^2 \rho^2 h^3 v^2}{C^2 P_0 k_B \Delta} \right) E^2 \tag{7}$

Here, the intrinsic loss tangent of the TLS is $\delta_{TLS}^0 = 1/Q_{TLS}^0$. Because $\bar{A}$ is proportional to the square of the electric field $E$ witnessed by the TLS, one can simply write $\bar{A} = \zeta \times E^2$ and $\zeta$ is a constant which is composed of $d_0, \rho, v, \gamma, P_0, C$, and $\Delta$, all of which are assumed to be electric field independent terms.

In addition to the $Q_{TLS}$ contribution to $Q_i$, the losses due to quasiparticles with density $n_{qp}$, including both non-equilibrium ($n_{non, eq}$) and equilibrium quasiparticles, should also be taken into consideration:

$$n_{qp} = n_{non, eq} + 2N_0 \sqrt{2\pi k_B T \Delta_{S0}} E \exp\left(-\frac{\Delta_{S0}}{k_B T}\right) \tag{8}$$

(valid for $T \ll T_c$) where $N_0 = 10^{47} J^{-1} m^{-3}$ (equivalent to $1.74 \times 10^4 \mu eV^{-1} \mu m^{-3}$\cite{7}\cite{13}) is the single spin density of states at the Fermi level, and hence $Q_{qp} \propto \frac{1}{n_{qp}}$, where $n_{qp}$ remains finite as $T \to 0$. In addition, there are some temperature independent loss mechanisms including moving vortices arising from stray magnetic field and geometry dependent microwave radiation loss, etc. The identity $\frac{1}{Q_{other}} = \frac{1}{Q_{vortices}} + \frac{1}{Q_{rad}}$ can be used to summarize these temperature independent loss mechanisms.

Therefore, one can say the internal quality factor of this half wavelength resonator follows the following expression in the limit $T \ll T_c$: $\frac{1}{Q_i} = \frac{1}{Q_{TLS}(T,E)} + \frac{1}{Q_{QP}(T,E)} + \frac{1}{Q_{other}}$ \tag{9}

In Fig. 4, the solid and dashed lines are generated by fitting the $Q_i(T)$ to the model described above. The quantities $n_{non, eq}, \Delta_{S0}, \bar{A}, Q_{TLS}^0, Q_{QP}(0K)$, and $Q_{other}$ are treated as fitting parameters. In addition, we take $\varepsilon = \hbar \omega_0$ for fitting the $Q_i(T)$. The fits give (see Table 1 in Sup. Mat. section II) $\Delta_{S0} = 185 \sim 188 \mu eV$, a gap value slightly larger than the value obtained from the frequency shift model above, due to the inclusion of non-equilibrium quasiparticles. This gap value is in agreement with many published experimental results of Al thin films\cite{52, 53}. Also, $n_{non, eq} \sim 50 \\mu m^{-3}$, a value which can be explained by theoretical treatment of the non-equilibrium distribution of quasiparticles and phonons in a superconductor driven by a continuous microwave photon flux\cite{6, 9, 11} based on our half wavelength CPW Al resonator geometry and experimental conditions (see supplemental material section III). This

FIG. 5. Applied electric field dependence of the fitting parameter $\bar{A}$ used to fit the $Q_i$ vs. temperature data at different circulating photon numbers. The dots are the extracted values from the fitting model, and the solid line is a fit to a quadratic formula with coefficient $\zeta = 3.49 \times 10^{-3} K \mu m^2/V^2$ and $b = 0.051 K$. The inset shows the same values plotted in a horizontal log scale to cover the higher electric field regime.
quasiparticle density is also consistent with the theoretical prediction \( n_{\text{non},\text{eq}} = 25 - 55 \mu m^{-3} \) for a similar type of Al thin film resonator.\([11]\) \( Q_{\text{others}} \) ranges from \( 1.43 \times 10^5 \) to \( 1.25 \times 10^5 \) and \( Q_{\text{TLS}}^0 \) also varies slightly from \( 1.7 \times 10^5 \) to \( 1.8 \times 10^5 \) for different circulating photon numbers, similar to the \( Q_{\text{max}} = 1/(F \times tanh) \) value found from fitting the frequency shift, above. These slight variations of \( Q_{\text{others}} \) and \( Q_{\text{TLS}}^0 \) could be due to the fitting errors. In summary, the non-equilibrium electron density \( n_{\text{non},\text{eq}} \) is found to be independent of photon circulation number at temperatures near the base temperature, allowing the TLS loss temperature dependence to dominate.

Due to the \( 1/T \) dependence of \( T_2 \) in this model, at low circulating photon numbers, the \( Q_i \) increases with decreasing temperature. This \( Q_i \) increase is suppressed when the excitation power is gradually increased, which saturates the TLS loss.

The fitting parameter, \( \tilde{A} \), in Eq. (7) is expected to change with photon number, and is shown in Fig. 5 as a function of applied electric field. The electric field witnessed by the TLS is estimated by scaling the Computer Simulation Technology Microwave Studio (CST) result of average electric field near the capacitive couplers under half watt excitation to the VNA output power by taking into account the attenuation in the input line with the assumption that the power is delivered to a 50 Ohm load impedance. These calculations are discussed in more detail in Supp. Mat. section IV. The solid line is a fit to the quadratic formula, \( \tilde{A} = \zeta E^2 + b \) with \( \zeta = 3.49 \times 10^{-3} K \cdot m^2/V^2 \) and \( b = 0.051 K \). In Supp. Mat. section V, we independently estimate the value of \( \zeta \) based on Eq. (7) and parameter values taken from glassy TLS systems, and find good agreement with the fit value. Clearly, the parameter \( \tilde{A} \) follows a square law on microwave electric field in the low power regime, consistent with the TLS model and the enhancement of TLS coherence at low temperatures.

\section*{V. DISCUSSION}

We also note that the fit of \( \tilde{A} \) vs. \( E \) in Fig. 5 starts to deviate from the quadratic curve at higher electric fields. This implies that at high applied power, the enhancement of \( Q_i \), due to the TLS model is interrupted by injected non-equilibrium quasiparticles and scattering between phonons and quasiparticles, as well as phonon trapping.\([8, 9, 54]\). A calculation of the increased quasiparticle density at high photon numbers in the half wavelength resonator can be found in Supp. Mat. section III. Note that this calculation includes the dynamics of the nonequilibrium quasiparticle finite lifetime due to recombination and trapping, with and without photon illumination\([44, 55]\). (Refer to Fig. S2 in the Supp. Mat.)

The upturn in \( Q_i(T) \) at low temperatures is due to an increase in TLS coherence times \( T_2 \sim 1/T \), in this model. Such an increase can come about due to interactions between the TLS.\([15, 50, 51]\) Evidence for interaction between TLS is well established from low temperature experiments on glasses.\([56, 57]\) Those studies have been interpreted as evidence of growing clusters of correlated TLS upon decreasing temperature.\([51]\) The TLS interact with each other by means of exchanging phonons, and can also be tuned by means of applied strain.\([58, 59]\) Direct measurements of \( T_1(T) \) and \( T_2(T) \) of individual TLS show significant enhancement for \( T < 300 \) mK,\([60]\) consistent with the upturn in \( Q_i(T) \) observed here.

\section*{VI. CONCLUSION}

We have designed and fabricated half wavelength superconducting aluminum microwave resonators with minimum critical dimension of 1 \( \mu m \) at the center conducting line of the CPW. The temperature dependence and microwave power dependence of the resonator \( Q_i \) shows an unusual increase with decreasing temperature when circulating microwave photon numbers are below \( 1 \times 10^4 \) and the frigde temperature is below 50 mK. These phenomena become more significant at circulating microwave photon numbers lower than several hundred. We attribute this behavior to the increase of TLS coherence time \( (T_2) \) at ultra-low temperature and power.

\section*{ACKNOWLEDGMENTS}

This work is funded by the US Department of Energy through Grant # DESC0018788. We acknowledge use of facilities at the Maryland Quantum Materials Center and the Maryland NanoCenter. We thank Rangga Budoyo and Braden Larsen for assistance with the nonequilibrium superconductor simulation. We thank Dr. Ben Palmer and Yizhou Huang at Laboratory for Physical Sciences for assistance with the thermal evaporator and helpful suggestions.

\begin{thebibliography}{99}
\bibitem{1} J. Zmuidzinas, Superconducting microresonators: Physics and applications, Annual Review of Condensed Matter Physics 3, 169 (2012).
\bibitem{2} J. J. A. Baselmans and S. J. C. Yates, Long quasiparticle lifetime in aluminum microwave kinetic inductance detectors using coaxial stray light filters, AIP Conference Proceedings 1185, 160 (2009).
\bibitem{3} P. Krantz, M. Kjaergaard, F. Yan, T. Orlando, S. Gustavsson, and W. Oliver, A quantum engineer’s guide to superconducting qubits, Applied Physics Reviews 6,
[4] M. Kjaergaard, M. E. Schwartz, J. Braumüller, P. Krantz, J. I.-J. Wang, S. Gustavsson, and W. D. Oliver, Superconducting qubits: Current state of play, Annual Review of Condensed Matter Physics 11, 369 (2020).

[5] M. D. Hutchings and et al, Tunable superconducting qubits with flux-independent coherence, Phys. Rev. Appl. 8, 1 (2017).

[6] J.-J. Chang and D. J. Scalapino, Kinetic-equation approach to nonequilibrium superconductivity, Physical Review B 15, 2651 (1977).

[7] D. J. Goldie and S. Withington, Non-equilibrium superconductivity in quantum-sensing superconducting resonators, Superconductor Science and Technology 26, 015004 (2013).

[8] P. J. de Visser, D. J. Goldie, P. Diener, S. Withington, J. J. A. Baselmans, and T. M. Klapwijk, Evidence of a nonequilibrium distribution of quasiparticles in the microwave response of a superconducting aluminum resonator, Physical Review Letters 112, 047004 (2014).

[9] R. P. Budoyo, Effects of Optical Illumination on Superconducting Quantum Devices, Ph.D. thesis, University of Maryland, College Park, USA (2015).

[10] A. P. Vepšaikänen, A. H. Karamlov, J. L. Orrell, A. S. Dogra, B. Loer, F. Vasconcelos, D. K. Kim, A. J. Melville, B. M. Niedzelski, J. L. Yoder, S. Gustavsson, J. A. Foraggio, B. A. VanDevender, and W. D. Oliver, Impact of ionizing radiation on superconducting qubit coherence, Nature 584, 551 (2020).

[11] P. J. de Visser, J. J. A. Baselmans, P. Diener, S. J. C. Yates, A. Endo, and T. M. Klapwijk, Number fluctuations of sparse quasiparticles in a superconductor, Physical Review Letters 106, 167004 (2011).

[12] R. Barends, J. Wenner, M. Lenander, Y. Chen, R. C. Bialczak, J. Kelly, E. Lucero, P. O’Malley, M. Mariantoni, D. Sank, H. Wang, T. C. White, Y. Yin, J. Zhao, A. N. Cleland, J. M. Martinis, and J. J. A. Baselmans, Minimizing quasiparticle generation from stray infrared light in superconducting quantum circuits, Applied Physics Letters 99, 113507 (2011).

[13] R. P. Budoyo, J. B. Hertzberg, C. J. Ballard, K. D. Voigt, J. R. A. Z. Kim, C. J. Lobb, and F. C. Wellstood, Effects of nonequilibrium quasiparticles in a thin-film superconducting microwave resonator under optical illumination, Phys. Rev. B 93, 024514 (2016).

[14] N. P. de Leon, K. M. Itoh, D. Kim, K. K. Mehta, T. E. Northup, H. Paik, B. S. Palmer, N. Samarth, S. Sangtawesin, and D. W. Steuerman, Materials challenges and opportunities for quantum computing hardware, Science 372, eabb2823 (2021).

[15] W. A. Phillips, Two-level states in glasses, Reports on Progress in Physics 50, 1657 (1987).

[16] J. Gao, J. Zmuidzinas, B. A. Mazin, H. G. LeDuc, and P. K. Day, Noise properties of superconducting coplanar waveguide microwave resonators, Applied Physics Letters 90, 102507 (2007).

[17] J. Gao, M. Daal, A. Vayonakis, S. Kumar, J. Zmuidzinas, B. Sadoulet, B. A. Mazin, P. K. Day, and H. G. Leduc, Experimental evidence for a surface distribution of two-level systems in superconducting lithographed microwave resonators, Applied Physics Letters 92, 152505 (2008).

[18] A. D. O’Connell, M. Ansmann, R. C. Bialczak, M. Hofheinz, N. Katz, E. Lucero, C. McKenney, M. Neely, H. Wang, E. M. Weig, A. N. Cleland, and J. M. Martinis, Microwave dielectric loss at single photon energies and millikelvin temperatures, Applied Physics Letters 92, 112903 (2008).

[19] S. Kumar, J. Gao, J. Zmuidzinas, B. A. Mazin, H. G. LeDuc, and P. K. Day, Temperature dependence of the frequency and noise of superconducting coplanar waveguide resonators, Applied Physics Letters 92, 123503 (2008).

[20] R. Barends, H. L. Hertingsius, T. Zijlstra, J. J. A. Baselmans, S. J. C. Yates, J. R. Gao, and T. M. Klapwijk, Contribution of dielectrics to frequency and noise of nbtin superconducting resonators, Applied Physics Letters 92, 223502 (2008).

[21] P. Makh, S. H. W. v. d. Ploeg, G. Oelsner, E. Il’ichev, H.-G. Meyer, S. Wünsch, and M. Siegel, Losses in coplanar waveguide resonators at millikelvin temperatures, Applied Physics Letters 96, 062503 (2010).

[22] C. Müller, J. H. Cole, and J. Lisenfeld, Towards understanding two-level-systems in amorphous solids: insights from quantum circuits, Reports on Progress in Physics 82, 124501 (2019).

[23] W. D. Oliver and P. B. Welander, Materials in superconducting quantum bits, MRS Bulletin 38, 816 (2013).

[24] A. Bruno, G. de Lange, S. Asaad, K. L. van der Enden, N. K. Langford, and L. DiCarlo, Reducing intrinsic loss in superconducting resonators by surface treatment and deep etching of silicon substrates, Appl. Phys. Lett. 106, 182601 (2015).

[25] G. Calusine, A. Melville, W. Woods, R. Das, C. Stull, V. Bolkhovsky, D. H. D. Brajel1, D. K. Kim, X. Miklósi, D. Rosenberg, A. Sevi, J. L. Yoder, E. Dauler, and W. D. Oliver, Analysis and mitigation of interface losses in trenched superconducting coplanar waveguide resonators, Appl. Phys. Lett. 112, 1 (2018).

[26] J. M. Sage, V. Bolkhovsky, W. D. Oliver, B. Turek, and P. B. Welander, Study of loss in superconducting coplanar waveguide resonators, J. Appl. Phys. 109, 063915 (2011).

[27] B. Chiaro, A. Megrant, A. Dunsworth, Z. Chen, R. Barends, B. Campbell, Y. Chen, A. Fowler, I. C. Hoi, E. Jeffrey, J. Kelly, J. Mutus, C. Neill, P. J. J. O’Malley, C. Quintana, P. Roushan, D. Sank, A. Vainsencher, J. Wenner, T. C. White1, and J. M. Martinis, Dielectric surface loss in superconducting resonators with flux-trapping holes, Supercond. Sci. Technol. 29, 104006 (2016).

[28] C. Richardson, A. Alexander, C. G. Weddle, B. Arey, and M. Olszta, Low-loss superconducting titanium nitride grown using plasma-assisted molecular beam epitaxy, J. Appl. Phys. 127, 235302 (2020).

[29] A. Romanenko, R. Pilipenko, S. Zorzetti, D. Frolov, M. Awida, S. Belomestnykh, S. Posen, and A. Grassellino, Three-dimensional superconducting resonators at t\(\alpha\)20 mk with photon lifetimes up to t\(\tau\)=2 s, Phys. Rev. Appl. 13, 034032 (2020).

[30] S. E. de Graaf, L. Faoro, L. B. Ioffe, S. Mahashabde, J. J. Burnett, T. Lindström, S. E. Kubatkin, A. V. Danilov, and A. Y. Tzalenchuk, Two-level systems in superconducting quantum devices due to trapped quasiparticles, Science Advances 6, eabc5055 (2020).

[31] C. Du, H. Chen, and S. Li, Quantum left-handed metamaterial from superconducting quantum-interference devices, Physical Review B 74, 113105 (2006).
L. Grünhaupt, N. Maleeva, S. T. Skacel, M. Calvo, S. J. Weber, K. W. Murch, D. H. Slichter, R. Vijay, and P. J. Petersan and S. M. Anlage, Measurement of resonance frequencies of microwave resonators, IEEE Transactions on Applied Superconductivity 21, 871 (2011).

C. R. H. McRae, H. Wang, J. Gao, M. R. Vissers, T. Brecht, A. Dunsworth, D. P. Pappas, and J. Mutus, Materials loss measurements using superconducting microwave resonators, Review of Scientific Instruments 91, 091101 (2020).

J. Burnett, L. Faoro, and T. Lindström, Analysis of high quality superconductor resonators: consequences for TLS properties in amorphous oxides, Superconductivity Science and Technology 29, 044008 (2016).

J. Gao, The Physics of Superconducting Microwave Resonators, PhD dissertation, Caltech, Department of Physics, Caltech (2008).

J. Burnett, L. Faoro, I. Wisby, V. L. Gurtovoi, A. V. Chernykh, G. M. Mikhailov, V. A. Tulin, R. Shaikhaidarov, V. Antonov, P. J. Meeson, A. Y. Tzalenchuk, and T. Lindström, Evidence for interacting two-level systems from the 1/f noise of a superconducting resonator, Nature Communications 5, 4119 (2014).

C. C. Yu and H. M. Carruzco, Two-level systems and the tunneling model: A critical view (2021), arXiv:2101.02787 [cond-mat.dis-nn].

A. E. Cherney, Enhancement of Superconductivity in Thin Aluminium Films, Master thesis, McMaster University (1969), a full PHDTHESIS entry.

N. A. Court, A. J. Ferguson, and R. G. Clark, Energy gap measurement of nanostructured aluminium thin films for single cooper-pair devices, Superconductor Science and Technology 21, 015013 (2007).

T. Guruswamy, D. J. Goldie, and S. Withington, Nonequilibrium superconducting thin films with sub-gap and pair-breaking photon illumination, Superconductor Science and Technology 28, 054002 (2015).

A. Rothwarf and B. N. Taylor, Measurement of recombination lifetimes in superconductors, Physical Review Letters 19, 27 (1967).

H. M. Carruzco, E. R. Grannan, and C. C. Yu, Nonequilibrium dielectric behavior in glasses at low temperatures: Evidence for interacting defects, Physical Review B 50, 6685 (1994).

H. M. Carruzco and C. C. Yu, Why phonon scattering in glasses is universally small at low temperatures, Physical Review Letters 124, 075902 (2020).

G. J. Grabovskij, T. Peichl, J. Lisenfeld, G. Weiss, and A. V. Ustinov, Strain tuning of individual atomic tunneling systems detected by a superconducting qubit, Science 338, 232 (2012).

J. Lisenfeld, G. J. Grabovskij, C. Müller, J. H. Cole, G. Weiss, and A. V. Ustinov, Observation of directly interacting coherent two-level systems in an amorphous material, Nature Communications 6, 6182 (2015).

J. Lisenfeld, C. Müller, J. H. Cole, P. Bushev, A. Lukashenko, A. Shnirman, and A. V. Ustinov, Measuring the temperature dependence of individual two-level systems by direct coherent control, Physical Review Letters 105, 230504 (2010).
Supplemental Material for Coherence Time Enhancement of Interacting Two-Level Systems in Aluminum Superconducting Resonators

Tamin Tai,1,* Jingnan Cai,1 and Steven M. Anlage1

1Quantum Materials Center, Department of Physics, University of Maryland, College Park, MD 20742-4111, USA

(Dated: September 24, 2021)

The Supplemental Material discusses the experimental details and measurement setup, gives the fitting parameters for the internal quality factor vs. temperature for different circulating photon numbers, and presents an overview of the nonequilibrium quasiparticle and phonon model used to describe the nonequilibrium quasiparticle loss in the resonator. An estimate of the parameter $\zeta$ based on microscopic properties of two-level systems (TLS) in glasses is also given.

I. EXPERIMENTAL DETAILS

Fig. S1 schematically shows the setup of the cryogenic transmission measurement with the VNA to characterize the resonator response. A variety of attenuators (produced by XMA) are used on each cryogenic stage to thermalize the center conductors of the coaxial cables. The total attenuation in the input line is -66 dB. Both the input line and output line on either side of the device are filtered by commercial microwave low-pass filters. The input line has a Marki Microwave low pass filter (FLP-1460) with 3-dB cutoff frequency at 14.6 GHz and the output line has another Marki Microwave low pass filter (FLP-1250) with 3-dB cutoff frequency at 12.5 GHz. The output line goes through the cryogenic isolator (QUINSTAR Technology QCI-G0301201AM) with working frequency band 3-12 GHz, and then the signal is amplified by 36 dB using a commercial high-electron mobility transistor amplifier (Low Noise Factory LNF-LNC0.3_14A with typical noise temperature 4.2 K) at the 4K stage, and then the transmitted signal is further amplified by 37 dB using another room temperature amplifier (HEMT) (Low Noise Factory LNF-LNC2.6A with typical noise temperature 50 K at ambient temperature).

* tamin@umd.edu
Fig. S1. Schematic of the microwave measurement setup for the study of the aluminum λ/2 resonator. The VNA at room temperature sends a signal from port 1 to the cryostat. The signal is attenuated at each stage of the cryostat before passing through the low-pass filter (LPF) and entering the device under test (DUT). The DUT is surrounded by a Cryoperm magnetic shield. The output signal also passes through a low-pass filter before going through 0-dB attenuators that thermalize the coaxial cable center conductor. The signal passes through an isolator and is amplified at the 4 K stage and at room temperature, before entering the VNA in port 2.

Table SI. Table of the model fitting parameters for the data in Fig. 4 of the main text, fit to Eq. (9), at different circulating photon numbers. $Q_{\text{TLS0}}$ and $Q_{\text{Other}}$ are the resonator quality factor coming from intrinsic dielectric loss tangent, and the quality factor coming from other loss mechanisms, respectively. The parameters $n_{\text{non,eq}}$, $Q_{\text{qp}}(0 \text{K})$ are the number density of non-equilibrium quasiparticles inside the aluminum layer and the quality factor affected only by the contribution of all (equilibrium and non-equilibrium) quasiparticles, respectively. The other fitting parameters are the superconducting energy gap ($\Delta_{\text{S0}}$) of the aluminum superconductor at 0 K and the coefficient $\tilde{A}$.

| photon number | $n_{\text{non,eq}}$ ($\mu$m$^{-3}$) | $Q_{\text{qp}}$ (0K) | $Q_{\text{TLS0}}$ | $Q_{\text{Other}}$ | $\Delta_{\text{S0}}$ (µeV) | $\tilde{A}$ (K) |
|---------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| $< n >$ 1     | 49.8 1.6 × 10^9 | 1.8 × 10^9      | 1.4 × 10^9      | 189 0.045       | 49.8 1.6 × 10^9 | 1.8 × 10^9      | 1.4 × 10^9      | 189 0.045       |
| $< n >$ 3     | 50.0 1.5 × 10^9 | 1.7 × 10^9      | 1.4 × 10^9      | 187 0.046       | 50.0 1.5 × 10^9 | 1.7 × 10^9      | 1.4 × 10^9      | 187 0.046       |
| $< n >$ 5     | 49.8 1.4 × 10^9 | 1.7 × 10^9      | 1.4 × 10^9      | 185 0.060       | 49.8 1.4 × 10^9 | 1.7 × 10^9      | 1.4 × 10^9      | 185 0.060       |
| $< n >$ 20    | 49.8 1.6 × 10^9 | 1.8 × 10^9      | 1.4 × 10^9      | 186 0.089       | 49.8 1.6 × 10^9 | 1.8 × 10^9      | 1.4 × 10^9      | 186 0.089       |
| $< n >$ 80    | 49.9 1.6 × 10^9 | 1.8 × 10^9      | 1.3 × 10^9      | 188 0.151       | 49.9 1.6 × 10^9 | 1.8 × 10^9      | 1.3 × 10^9      | 188 0.151       |
| $< n >$ 325   | 49.9 1.4 × 10^9 | 1.8 × 10^9      | 1.3 × 10^9      | 188 0.399       | 49.9 1.4 × 10^9 | 1.8 × 10^9      | 1.3 × 10^9      | 188 0.399       |
| $< n >$ 1 × 10^3 | 50.0 1.4 × 10^9 | 1.8 × 10^9      | 1.3 × 10^9      | 188 1.636       | 50.0 1.4 × 10^9 | 1.8 × 10^9      | 1.3 × 10^9      | 188 1.636       |
| $< n >$ 1 × 10^4 | 49.9 1.4 × 10^9 | 1.9 × 10^9      | 1.3 × 10^9      | 187 3.188       | 49.9 1.4 × 10^9 | 1.9 × 10^9      | 1.3 × 10^9      | 187 3.188       |

II. FITTING PARAMETERS FOR QUALITY FACTOR VS. TEMPERATURE

Table SI lists the fitting parameters used to fit the internal quality factor vs. temperature data for different circulating photon numbers, shown as solid and dashed lines in Fig. 4 of the main text. Note that most of the systematic variation with circulating photon number is accommodated by $\tilde{A}$. 
III. NON-EQUILIBRIUM QUASIPARTICLE TREATMENT AND n_{qp} AND Q_{qp} ESTIMATES

The aim of this work is to independently examine the non-equilibrium quasiparticle density fitting parameter listed in Table S1 by simulating the physics using the non-equilibrium energy-dependent distribution of quasiparticles. This model (scattering model) was established by considering the coupled quasiparticle and phonon systems [1]. The details of this model are described in references [2] [3] and [4] and employ numerical methods to discretize the distribution of quasiparticles $f(E)$ at energy $E$ and the phonon distribution $n(\Omega)$ at energy $\Omega$ by solving the kinetic equations in steady state for a given flux of microwave photons and higher frequency radiation.

![Fig. S2](image-url)

Fig. S2. The calculated quasiparticle distribution $f(E)$ (a) and phonon distribution $n(\Omega)$ (b) as a function of normalized energy for different circulating numbers of photons in the half wavelength resonator. Note that in the plot of $f(E)$, a double y axis is used due to the different scales of thermal and non-equilibrium distributions.

Fig. S2 (a) and Fig. S2 (b) show the calculated quasiparticle distribution and phonon distribution, respectively, for three selected circulated numbers of photons in the half wavelength resonator. This calculation is performed by assuming the fridge base temperature $T_b = 10$ mK, the resonator drive frequency 3.6442 GHz ($\hbar\omega = 23 \mu eV$), superconducting energy gap $\Delta_s = 188 \mu eV$, $Q_i = 10^5$, $Q_c = 1.5 \times 10^6$ which we obtain from fitting the resonance, resonator center conductor volume $8.6 \times 10^{-14}$ m$^3$ and effective temperature $T_{eff} = 189$ mK due to stray light illumination and radiation which creates enhanced phonon generation, as described by the Parker model [5]. Table SII lists all of the parameter values used in this model. The black dashed line in Fig. S2(a) indicates the thermal distribution of quasiparticles without any microwave excitation at $T_b = 10$ mK. At low circulating photon numbers ($< n > = 1$ or 325), the quasiparticle distribution is enhanced significantly above the $T_b$ thermal distribution, and the phonon distribution is also enhanced. At high circulating photon numbers, jumps appear in the electron and phonon distributions every $\hbar \omega$ because microwave drive at high power significantly affects the distributions. Big step jumps at $E = 3\Delta_s$ and $\Omega = 2\Delta_s$ are due to pair breaking and recombination processes.

From the quasiparticle distribution $f(E)$, one can calculate the quasiparticle density, $n_{qp}$, by numerical integration over all energy, with its density of states $\rho(E)$

$$n_{qp} = 4N_0 \int_{\Delta_s}^{\infty} f(E)\rho(E)dE \quad \text{with} \quad \rho(E) = \frac{E}{\sqrt{E^2 - \Delta_s^2}}$$

(S1)

The calculated $n_{qp}$ at different circulated photon numbers $< n >$ are shown in Fig. S3(a). Because this calculation is performed at the 10 mK fridge base temperature, the entire term $n_{qp}$ (Eq. (8) of the main text) can be regarded as the contribution of non-equilibrium quasi-particle density since the number of thermal quasiparticles is extremely small. The calculated $n_{qp} \sim 50 \mu m^{-3}$ when $< n > < 10^6$ is consistent with the parameter value $n_{non, eq}$ in Table I from fitting the $Q_i(T)$ by using the model described in the main text. In addition, the calculated result of $n_{qp}$ as a function of fridge/bath temperature ($T_b$) at $< n > = 1$ is shown in Fig. S3(b) as circle dots. When the bath temperature is below 150 mK, $n_{qp}$ remains constant. This is consistent to the assumptions made in the frequency shift fit at low temperature where quasiparticle density is a constant. The continuous line of Fig. S3(b) is from Eq. 8 of the main text, made by this assumption.
Superconducting gap ($\Delta_{s0}$) = 188 $\mu$eV

$Q_i = 1.0 \times 10^5$

Quasiparticle-phonon time = 438 ns [3]

Phonon escape time = 0.17 ns [3]

Base Temperature $T_b$ = 10 mK

RF photon energy ($h\omega$) = 23 $\mu$eV

$Q_e = 1.5 \times 10^6$

Characteristic phonon time = 0.26 ns [3]

Resonator volume = 8.6 $\times 10^{-14}$ m$^3$

Phonon effective temperature ($T_{eff}$) = 189 mK

| Parameter | Value |
|-----------|-------|
| $\Delta_{s0}$ | 188 $\mu$eV |
| $Q_i$ | $1.0 \times 10^5$ |
| Quasiparticle-phonon time | 438 ns |
| Phonon escape time | 0.17 ns |
| Base Temperature $T_b$ | 10 mK |
| $Q_e$ | $1.5 \times 10^6$ |
| Characteristic phonon time | 0.26 ns |
| Resonator volume | 8.6 $\times 10^{-14}$ m$^3$ |
| Phonon effective temperature $T_{eff}$ | 189 mK |

Table SII. Parameters used for our non-equilibrium quasi-particle calculation.

For the quality factor due to the quasiparticles, $Q_{qp}$ is defined as

$$\frac{1}{Q_{qp}} = \alpha \frac{\sigma_1}{\sigma_2}$$

where the kinetic inductance ratio $\alpha = 0.0185$ was obtained from the frequency shift fit in the main text. The real and imaginary parts of the complex conductivity $\sigma = \sigma_1 - j\sigma_2$ can be expressed respectively by the Mattis-Bardeen formula [6] given by

$$\frac{\sigma_1}{\sigma_N}(\omega) = \frac{2}{\hbar\omega} \int_0^\infty [f(E) - f(E + h\omega)] g_1(E) dE$$

$$\frac{\sigma_2}{\sigma_N}(\omega) = \frac{2}{\hbar\omega} \int_0^\infty [f(E) - 2f(E + h\omega)] g_2(E) dE$$

where $\sigma_N$ is the normal-state conductivity, $g_1(E) = h_1(E, E + h\omega)\rho(E)$ and $g_2(E) = h_1(E, E + h\omega)\frac{E}{\sqrt{\Delta_{s0}^2 - E^2}}$ with $h_1(E, E') = (1 + \frac{\Delta_{s0}^2}{E^2})\rho(E')$. Here we use the non-equilibrium distribution function $f(E)$ discussed above.

The calculated result of $Q_{qp}$ as a function of average circulated photon numbers is shown in Fig. S4. In the regime of low circulating photon numbers, $Q_{qp}$ remains a constant and then gradually increases as the circulating photon numbers in the half wavelength resonator increase. Overall, the $Q_{qp}$ is on the order of $10^7$, close to the values in Table SI, and this result verifies our assumption that $Q_{qp} > Q_{TLS0}$ in the $Q_i(T)$ fitting. In other words, the loss in the half wavelength resonator at low temperatures and low circulating photon numbers is dominated by the TLS losses.

Fig. S3. (a) The calculated quasiparticle density as a function of circulating photon numbers in the half wavelength resonator. Here $T_b = 10 mK$ and $T_{eff} = 189 mK$. (b) The calculated quasiparticle density as a function of fridge temperature at $<n> = 1$. Note the dots are calculated from the non-equilibrium model, and the solid line is from Eq. 8 of the main text.
Fig. S4. The calculated quality factor of quasi-particle as a function of circulating photon numbers \(< n >\) in the half wavelength resonator. Here \(T_b = 10\) mK and \(T_{eff} = 189\) mK, and other parameters are listed in Table SII.

IV. CST MICROWAVE SIMULATION OF CPW ELECTRIC FIELDS

A model of the CPW microwave resonator was constructed in CST Microwave Studio. The model structure (Fig. S5) represents the entire CPW resonator and coupling capacitors, both of which reproduce the geometrical structure in our superconducting chip. The superconductor is modeled as a perfect electric conductor for the purpose of E field calculation. The results below are obtained from the finite element frequency domain solver in CST.

The CST simulation shows that on resonance at 3.647 GHz, the electric field can attain a maximum of \(6 \times 10^8\) V/m on the substrate-vacuum interface at the corner of the center strip that is part of the coupling capacitor on the input side. This calculation was done under a 0.5 watt excitation level. An average electric field of the adjacent area is estimated to be \(\sim 1.16 \times 10^8\) V/m. By scaling this power down to that required to achieve one circulating photon in the resonator, we estimate the average electric field of the region nearby the coupling capacitor of the resonator to be \(0.2\) V/m. In Fig. 5 of the main text, the E field on the x-axis is estimated from this scaling and extended to the other circulating photon numbers in the resonator.

V. TWO-LEVEL SYSTEM MODEL PARAMETER ESTIMATES AND COMPARISON TO DATA

Here we estimate from first principles the magnitude of \(\tilde{A} = \zeta \times E^2\) (Eq. (7) of the main text) and compare to the values derived from fits to the \(\tilde{A}\) vs. electric field experimental data shown in Fig. 5 of the main text.

\[
\tilde{A} = \left(\frac{4d_0^2}{3\varepsilon^2} \left[ \frac{\gamma_L^2}{v_L^2} + \frac{\gamma_T^2}{v_T^2} \right]^{-1} \frac{\pi^2 \rho^2 k^2 v^2}{C\gamma^2 P_0 k_B \Delta} \right) E^2 \equiv \zeta E^2
\]  

(S5)

We make use of the extensive literature on TLS in glasses at low temperatures, as well as the more recent literature on individual TLS spectroscopy. It is observed experimentally that the average properties of TLS in glass are essentially universal,[7] hence in that spirit we use values of the parameters that are gathered from different glasses. This calculation is intended as no more than an order of magnitude estimate to see if the experimental value for \(\zeta\) is roughly on the scale of what we expect from widely accepted microscopic parameters.

The parameters used to estimate \(\zeta\) are shown in Table SIII, along with the source for each value. We note that the recent experiments on single TLS spectroscopy extract values for these parameters that are close to those from the low-temperature glass literature. For example, values of the deformation potential for aluminum oxide of \(\gamma \approx 0.2 - 0.4\) eV are extracted from single TLS measurements,[8] a bit lower than the value of \(\gamma\) obtained from measurements of fused silica glasses.[9]

Using the numbers in Table SIII we find that \(\zeta_{estimated} = 2.6 \times 10^{-3}\) K \(\text{m}^2/\text{V}^2\). This is close to the experimental value extracted from the fit to the data in Fig. 5 of the main text, namely \(\zeta_{exp} = 3.49 \times 10^{-3}\) K \(\text{m}^2/\text{V}^2\). Hence we believe that the experimentally obtained value for \(\zeta\) is reasonable.
Fig. S5. CPW resonator model in CST microwave studio. (a) The geometry of the CPW resonator model in CST. The coupling capacitors and the entire resonator structure reproduce those in the experiment. (b) The top view of the E field strength on the substrate-vacuum interface at the fundamental resonance of 3.647 GHz, which has a node in the center of the resonator, a typical standing wave pattern in a half wavelength resonator. (c) The close-up top view of the E field vector plot on the substrate-vacuum interface near the coupling capacitor. The maximum E field is found around the corner of the center strip that is part of the coupling capacitor. The E field that contributes to the TLS model is estimated from the average of the field along the resonator side of the coupler about $1.16 \times 10^8 \text{V/m}$, highlighted in the yellow-orange line. (d) The side view of the E field vector plot on the cross section through the coupler at the center strip corner. The maximum is at the corner of the center strip on the substrate-vacuum interface.

[1] J.-J. Chang and D. J. Scalapino, Kinetic-equation approach to nonequilibrium superconductivity, Physical Review B 15, 2651 (1977).
[2] D. J. Goldie and S. Withington, Non-equilibrium superconductivity in quantum-sensing superconducting resonators, Superconductor Science and Technology 26, 015004 (2013).
[3] P. J. de Visser, D. J. Goldie, P. Diener, S. Withington, J. J. A. Baselmans, and T. M. Klapwijk, Evidence of a nonequilibrium distribution of quasiparticles in the microwave response of a superconducting aluminum resonator, Physical Review Letters 112, 047004 (2014).
[4] R. P. Budoyo, Effects of Optical Illumination on Superconducting Quantum Devices, Ph.D. thesis, University of Maryland, College Park, USA (2015).
[5] W. H. Parker, Modified heating theory of nonequilibrium superconductor, Physical Review B 12, 3667 (1975).
[6] D. C. Mattis and J. Bardeen, Theory of the anomalous skin effect in normal and superconducting metals, Physical Review 111, 412 (1958).
[7] C. C. Yu and H. M. Carruzo, Two-level systems and the tunneling model: A critical view (2021), arXiv:2101.02787 [cond-mat.dis-nn].
[8] G. J. Grabovskij, T. Peichl, J. Lisenfeld, G. Weiss, and A. V. Ustinov, Strain tuning of individual atomic tunneling systems detected by a superconducting qubit, Science 338, 232 (2012).
[9] J. L. Black, Relationship between the time-dependent specific heat and the ultrasonic properties of glasses at low temperatures, Physical Review B 17, 2740 (1978).
[10] A. M. Holder, K. D. Osborn, C. J. Lobb, and C. B. Musgrave, Bulk and surface tunneling hydrogen defects in alumina, Physical Review Letters 111, 065901 (2013).
[11] B. Sarabi, A. N. Ramanayaka, A. L. Buin, F. C. Wellstood, and K. D. Osborn, Projected dipole moments
| Parameter                                      | Symbol | Value and units          | Comments and Reference                                                                 |
|-----------------------------------------------|--------|--------------------------|----------------------------------------------------------------------------------------|
| Transition dipole moment                      | $d_0$  | $1 \text{ e} - \text{Å}$ | Order of magnitude estimate based on Refs. [10],[11]                                   |
| TLS energy level splitting                    | $\varepsilon$ | $hf = 2.41 \times 10^{-24} \text{J}$ | Energy level splitting matches photon energy                                            |
| TLS well offset energy                        | $\Delta$ | $hf = 2.41 \times 10^{-24} \text{J}$ | Energy level asymmetry on the scale of photon energy                                   |
| TLS DOS*Longitudinal Deformation Potential    | $\rho L_0$ | $1.4 \times 10^7 \text{ J/m}^3$ | Fused silica value [9]                                                               |
| TLS DOS*Transverse Deformation Potential      | $\rho T_0$ | $0.63 \times 10^7 \text{ J/m}^3$ | Fused silica value [9]                                                               |
| Longitudinal Sound Velocity                   | $v_L$  | $5.8 \times 10^3 \text{ m/s}$ | Fused silica value [9]                                                               |
| Transverse Sound Velocity                     | $v_T$  | $3.8 \times 10^3 \text{ m/s}$ | Fused silica value [9]                                                               |
| Mass Density                                  | $\rho$ | $2.2 \times 10^3 \text{ kg/m}^3$ | Fused silica value [9]                                                               |
| Averaged Sound Velocity                       | $v$    | $4.16 \times 10^3 \text{ m/s}$ | Assuming $1/v^3 = \frac{1}{3} \sum_{r=1}^{3} \frac{1}{v_i^3}$ [7]                   |
| Dimensionless Phonon scattering rate [7]      | $C$    | 1                        | Reference [12]                                                                       |
| Deformation Potential                         | $\gamma$ | $1.6 \text{ eV}$ | Fused silica value [9]                                                               |
| Electric Field at Single-Photon Level         | $E$    | $0.22 \text{ V/m}$ | Estimated from CPW full-wave simulation                                               |

Table SIII. Table of estimated parameters that go into a calculation of the quantity $\tilde{A} = \zeta E^2$, Eq. (7) of the main text. A representative value of each parameter is chosen for use in an order of magnitude estimate of $\zeta$.

DOS = density of states.

of individual two-level defects extracted using circuit quantum electrodynamics, Physical Review Letters 116, 167002 (2016).

[12] C. Yu and A. Leggett, Low temperature properties of amorphous materials: Through a glass darkly, Comments Cond. Mat. Phys 14, 231 (1988).