Three-Dimensional Superconducting Resonators at $T < 20$ mK with Photon Lifetimes up to $\tau = 2$ s

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Very-high-quality-factor superconducting radio-frequency cavities developed for accelerators can enable fundamental physics searches with orders of magnitude higher sensitivity, and they can also offer a path to a 1000-fold increase in the achievable coherence times for cavity-stored quantum states in three-dimensional circuit QED architecture. Here we report measurements of multiple accelerator cavities of resonant frequencies of $f_0 = 1.3$, 2.6, 5 GHz down to temperatures of about 10 mK and field levels down to a few photons, which reveal very long photon lifetimes up to 2 s, while also further exposing the role of the two-level systems (TLS) in niobium oxide. We also demonstrate how the TLS contribution can be greatly suppressed by vacuum heat treatments at 340–450 °C.

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I. INTRODUCTION

Superconducting radio-frequency (SRF) cavities in particle accelerators routinely achieve [1,2] very high quality factors $Q > 10^{10}$–10^{11} corresponding to photon lifetimes τ as long as tens of seconds. These are much higher than the highest $Q \sim 10^8$ reported in various quantum regime studies [3,4] with $\tau \sim 1$ ms. Thus, adopting SRF cavities for a three-dimensional (3D) circuit QED architecture for quantum computing or memory appears to be a very promising approach due to the potential of a 1000-fold increase in the photon lifetime and therefore cavity-stored quantum state coherence times. There is also a variety of proposed fundamental physics experiments, i.e., dark photon and axion searches [5–7], for which the availability of higher Q cavities in the lower-photon-count regime would directly translate into multiple orders of magnitude increases in search sensitivities.

Recent investigations [8] have revealed that the two-level systems (TLS) residing inside niobium oxide may play a significant role in the low-field performance of SRF cavities, similar to two-dimensional (2D) resonators [9,10]. To gain further understanding of the physics involved, and to guide future Q improvement directions, a direct probing of SRF cavities in the quantum regime is required.

In this article, we report measurements of a selection of state-of-the-art SRF cavities down to very low temperatures ($T < 20$ mK) and very low fields of a few photons (the "quantum" regime). We achieve very long photon lifetimes of more than 2 s, and observe a Q decrease when going from previously explored temperatures of 1.3 K or above down to below 20 mK. This is also a direct study of the TLS in 3D Nb resonators in the quantum regime, as well as a demonstration of the drastic TLS-induced dissipation decrease associated with the oxide removal. Our results demonstrate that SRF cavities can serve as a very long coherence platform for, e.g., 3D circuit QED and quantum memory [4,11] applications, as well as for various fundamental physics experiments, such as dark photon or axion searches [5–7].

II. EXPERIMENTAL APPROACH

We use single-cell niobium cavities of the TESLA shape [12] with resonant frequencies f_0 of the TM_{010} modes of 1.3, 2.6, and 5.0 GHz, made of fine-grain bulk niobium with high residual resistivity ratio of $\gtrsim 200$.

The cavities utilized are shown in Fig. 1 along with the calculated electric and magnetic field distributions. The fundamental frequency sets the radial dimension R of the cavities: $R \propto 1/f_0$. Electromagnetic coupling to the cavities is performed using axial pin couplers at both ends of the beam tubes.

We also apply targeted heat treatments in a custom-designed furnace [13] to remove the niobium pentoxide

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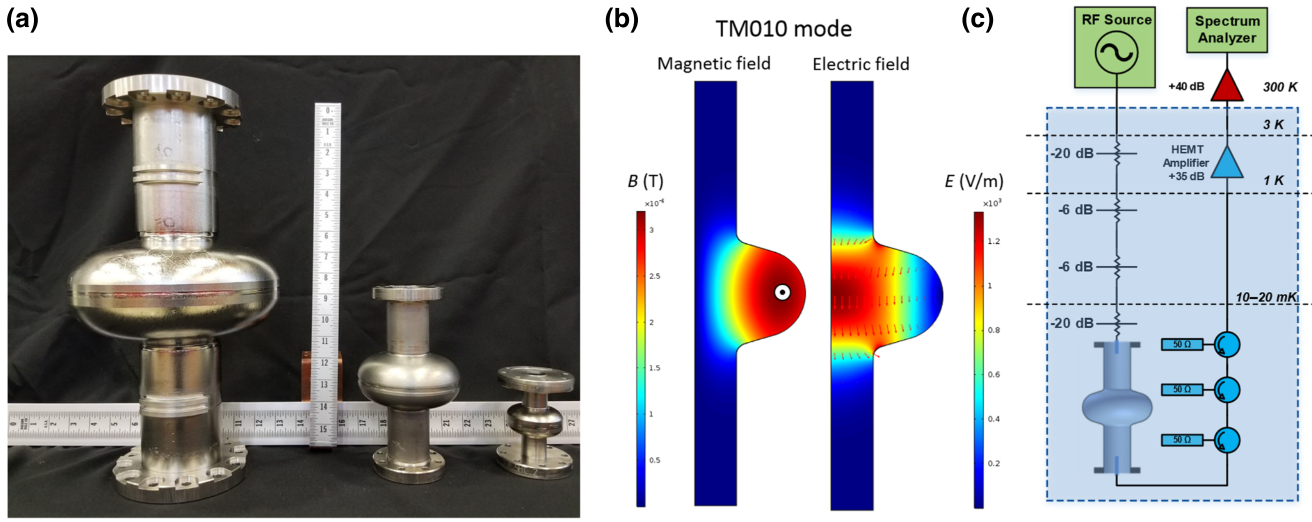

FIG. 1. (a) Single-cell cavities of the TESLA geometry used for the measurements. (b) Distributions of the magnetic and electric fields in the TM010 mode (half of the rotationally symmetric cavity is shown); the coupling to the mode is performed using pin couplers on the cavity axis on both sides. (c) The typical microwave setup used for measurements (the attenuators and the amplifiers are different in some cases for different frequencies and cooldown cycles).

(Nb_2O_5) and to directly investigate the associated improvement in the TLS dissipation on both the 1.3-GHz and 5-GHz cavities. The 1.3-GHz cavity is heat treated at 340°C in vacuum for several hours, whereas the 5-GHz cavity is treated similarly at 450°C as the last step of the cavity preparation.

Measurements are performed first at the vertical test facility where the cavities are submerged in liquid helium and temperatures down to 1.4 K can be achieved, and then in the dilution refrigerator at temperatures down to 10–20 mK.

For cavities in the vertical test dewar we use the standard SRF measurement techniques [14] at higher accelerating fields E, and the filtered decay method [8] at lower fields. The $Q(E)$ results at temperatures down to 1.4 K in a broad range of higher cavity fields are shown in Fig. 2. It is notable that the 1.3-GHz cavity after the 340°C heat treatment has an extremely high quality factor $Q \gtrsim 4 \times 10^{11}$ in a broad range of fields, higher than in e.g. [2]. This indicates that the 340°C heat treatment suppresses the residual resistance at all fields, which may also be related to the TLS or other potential mechanisms, i.e., to the elimination of the possible metallic niobium suboxide inclusions inside the pentoxide layer.

For dilution refrigerator measurements, a double-layer magnetic shielding around the full cryostat is used, and magnetometers placed directly on the outside cavity surfaces indicate that the dc ambient magnetic field level is shielded to below 2 mG in all cases. The microwave setup includes a series of attenuators on the cavity input line, as well as both cryogenic and room-temperature amplifiers on the pickup line. For one of the runs with the 5-GHz cavity, the Josephson parametric amplifier (JPA) is used in the output line as well. The measurement configuration including the low-noise cryogenic amplifier (HEMT) makes it possible to measure reliably the photon lifetimes down to an average cavity population of $\overline{n} \sim 10$ photons, while JPA extends the sensitivity further to single-photon levels. The cavity placement and a typical microwave schematic of the setup are shown in Fig. 1(c).

The average photon number is calculated from the cavity stored energy U: $\overline{n} = U/\hbar\omega$, where $U = P_t Q_t/\omega$ is

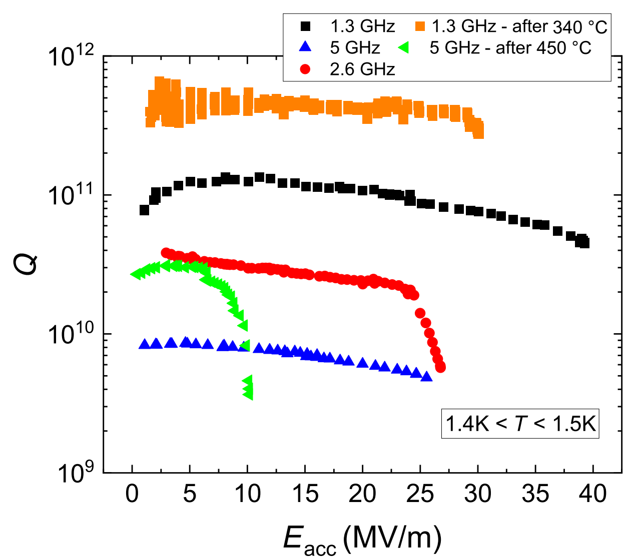

FIG. 2. Intrinsic cavity quality factors of the investigated 1.3-, 2.6-, and 5-GHz cavities as a function of the accelerating field at temperatures $1.4 < T < 1.5$ K.

extracted from the measured transmitted signal P_t at the pickup coupler with the external quality factor Q_t.

III. RESULTS

A. Cavity measurements

Typical decay curves for 5-GHz cavities before (blue and black curves) and after (magenta and dark yellow curves) 450°C vacuum heat treatment for different starting cavity photon populations are shown in Fig. 3. For each case, decays from two different starting power levels are shown, corresponding to two different resolution bandwidths of the spectrum analyzer as well. As the exponential decay fits (red lines) indicate, the time constant at different stored energy levels and therefore the quality factor Q remains constant down to the noise floor of about 10 photons and approximately 2 photons, respectively. We also observe no rf field-amplitude dependence of the Q factor for all the cavities in the dilution refrigerator setup. This is consistent with our previous studies and higher-temperature and higher-field measurements in the current study, which showed that the "critical" TLS saturation field E_c for niobium oxide is much higher—of the order of $E_c \sim 0.1$ MV/m—and therefore TLS are not saturated by the microwave fields from about $\bar{n} \sim 10^{20}$ all the way down to $\bar{n} \sim 2$.

$Q(T)$ measurements, which represent the main findings of our paper, are shown in Fig. 4. A characteristic $\propto 1/\tanh[\alpha(\hbar\omega/2kT)]$ temperature dependence of the quality factors $Q(T)$ for all the cavities is clearly observed with the Q decreasing towards lower temperatures. The amount of Q degradation is drastically suppressed by the heat treatments—340°C for the 1.3-GHz cavity and 450°C for the 5-GHz cavity, respectively. This is consistent with the removal of the significant number of TLS, which are hosted by the pentoxide layer of SRF cavities, as shown in our previous work [8].

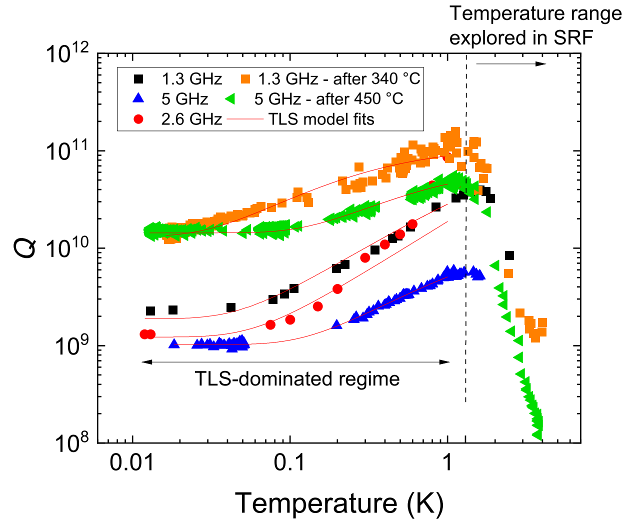

FIG. 4. Intrinsic cavity quality factor Q of the 1.3-, 2.6-, and 5-GHz cavities as a function of temperature. Previously, SRF cavities have been studied at temperatures above about 1.3 K. Below about 1 K a significant decrease in Q is observed consistent with the TLS dissipation; the red lines show TLS model fits. A dramatic increase in Q associated with the oxide-modifying heat treatment is apparent on the 1.3-GHz and 5-GHz cavities.

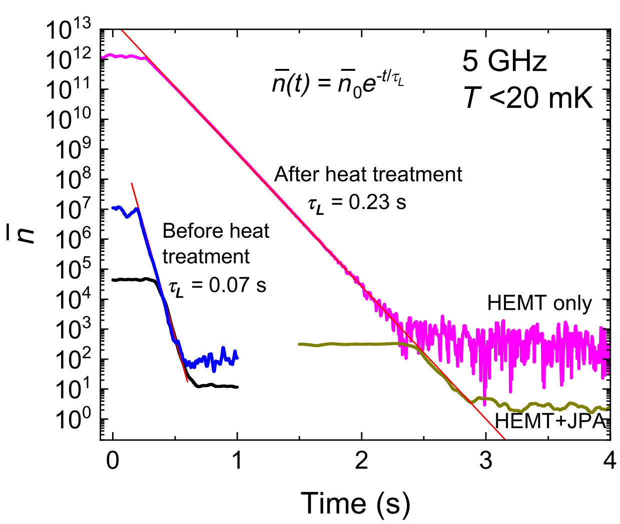

FIG. 3. Average photon population decay upon switching the rf power off measured in the 5-GHz cavities before and after heat treatment at 450°C. Blue and black curves correspond to the decays from different starting fields and with different filtering bandwidths (100 Hz and 10 Hz, respectively). The red lines show linear fits for both. The magenta and dark yellow lines show stored energy decays of the 450°C treated cavity with the much longer photon lifetime.

Below about 1 K the contribution to the surface resistance caused by thermally excited quasiparticles becomes negligible and the $Q(T)$ curves appear to be dominated by dissipation caused by the TLS. The TLS dissipation increases as the temperature is further lowered due to decreased thermal saturation and therefore an increased number of TLS systems participating in the resonant absorption of the microwave power.

B. TLS model fitting

In the TLS-dominated regime, an excellent fit is obtained using the "standard" TLS model [9,10] dissipation, with δ_0 as the loss tangent of the TLS at $T = 0$ K, an additional coefficient α to account for temperature measurement efficiency, and a fixed residual (R_{res}) surface resistance:

$$\frac{1}{Q(T)} = F\delta_0 \tanh\left(\alpha\frac{\hbar\omega}{2kT}\right) + \frac{R_{\text{res}}}{G}, \quad (1)$$

TABLE I. Summary of TLS model fitting results.

f_0 (GHz)	Oxide treatment	$F\delta_0$	F	δ_0
1.3	No	5.2×10^{-10}	1.0×10^{-7}	0.17
2.6	No	8.2×10^{-10}	2.4×10^{-8}	0.13
5	No	9.1×10^{-10}	1.2×10^{-8}	0.08
1.3	340°C 3 h	6.7×10^{-11}	...	...
5	450°C 3 h	5.6×10^{-11}	...	...

where F is the calculated filling factor [4,15,16], $G = 268\ \Omega$ is the geometry factor of the TM_{010} mode for the TESLA shape extracted from the finite-element simulations [12].

In Table I the $F\delta_0$ values from the obtained fit are shown for all the cavities investigated. A dramatic—about an order of magnitude—decrease in $F\delta_0$ is associated with the heat treatments. For cavities before the heat treatments (with the pentoxide layer) the participation ratios can be calculated as in Ref. [8] assuming an oxide layer of approximately 5 nm, and the δ_0 values can then be estimated as well. These are listed in Table I wherever applicable.

While the presence of TLS leads to a decrease of Q from its values at higher temperatures, even in the worst case (5 GHz without heat treatment) we obtain a photon lifetime $\tau = 32$ ms, which is several times higher than that previously reported of approximately 10.2 ms [17]. After heat treatment, the achieved photon lifetimes of 0.5–2 s correspond to an improvement of approximately 50–200 times.

C. Time-of-flight secondary ion mass spectrometry surface studies

To reveal the underlying material changes happening during the vacuum treatment in the temperature range of interest (340–450°C), we perform direct studies on the niobium cavity cutout using an in-house time-of-flight secondary ion mass spectrometry (TOF-SIMS) system. TOF SIMS measures the depth profiles of various elements within the sample with subnanometer depth resolution and better than parts-per-million concentration resolution, and has been actively used in recent years to guide the tailoring of SRF cavity near-surface structure [18]. Shown in Fig. 5, the comparison before and after the 400°C vacuum treatment (without subsequent air exposure) confirms the removal of the Nb_2O_5, likely explaining the reduced TLS dissipation after $340 - 450$°C treatments. Furthermore, we discover the emergence of a strong near-surface nitrogen enrichment, which is the likely cause of the "doping"-like effect we find at higher cavity fields after these heat treatments [19].

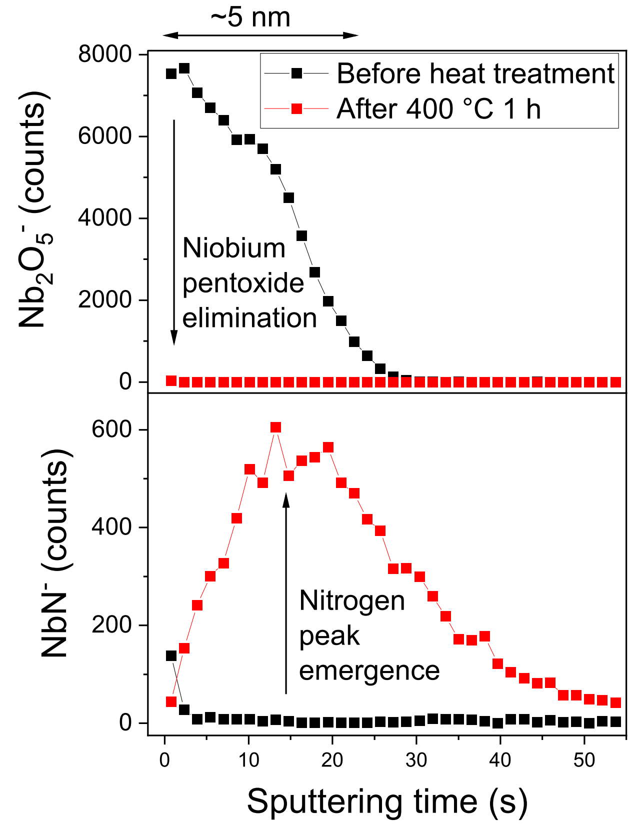

FIG. 5. TOF-SIMS depth profiles obtained on the cavity cutout revealing the changes in the niobium pentoxide and nitrogen-related signals after 400°C *in situ* heat treatment. About a 5-nm-thick Nb_2O_5 pentoxide layer is completely dissolved after the 400°C treatment. Another apparent change is the increase in the nitrogen level within approximately 10 nm of the surface.

IV. DISCUSSION

It is intriguing that the cavities after the $340 - 450$°C heat treatments still have some nonzero Q degradation at temperatures lower than 1 K (Fig. 4). Since SIMS studies suggest that there is no Nb_2O_5 after these treatments, an additional source of TLS should be present as well. Some potential examples could be other types of niobium oxides (e.g., NbO) and their interfaces with the underlying bulk, or, e.g., surface adsorbates. Pinpointing these remaining sources would be a key goal of future detailed investigations.

In practise, our findings open up a pathway to explore coupled SRF cavity-transmon structures as the highest coherence superconducting quantum circuits for quantum computing. In particular, implementing the protocol from Ref. [20] would allow direct generation of very long-lived Fock states in SRF cavities. One important question is what is the best way to insert the transmon in the SRF cavity to provide enough coupling to the cavity mode of interest while not degrading the ultra-high Q. A possible solution is the use of a low-loss dielectric rod (such as sapphire) to hold the transmon in the relevant field area of the cavity. Corresponding electromagnetic design work and the mechanical and microwave measurements to validate this concept are currently under way.

For new physics searches, using SRF cavities with the Q factors we demonstrate, allow for a sensitivity increase of multiple orders of magnitude. A prototype dark-photon "light-shining-through-the-wall" search experiment of the type in Ref. [5] but now with much higher Q SRF cavities has been assembled and is currently being commissioned with results to be reported in future publications.

V. CONCLUSION

In summary, we perform measurements of state-of-the-art SRF accelerator cavities in the quantum regime and demonstrate photon lifetimes as high as $\tau = 2$ s—about a factor of 200 higher than other results in this regime. We also reveal a quality factor decrease at lower temperatures, consistent with the contribution of the TLS hosted by the niobium oxide, and demonstrate its mitigation by *in situ* heat treatments at $340 - 450°$C resulting in the removal of niobium pentoxide, as witnessed by TOF SIMS.

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