

Nonequilibrium dynamics of two-level systems directly after cryogenic alternating bias

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Two-level systems (TLSs) are a dominant decoherence mechanism in superconducting qubits, microwave kinetic inductance detectors, and quantum dots. TLSs are tunneling states commonly found in amorphous materials and interfaces, which interact electromechanically, leading to energy loss. Fluctuations in the frequency of the TLSs are a significant problem for qubit operation and force recalibration every few hours. In this study, we probe the effect of alternating bias at cryogenic temperatures on TLSs in a purpose-built LC oscillator. When an *in situ* alternating bias is applied, the steady-state spectra of the TLSs disappear. Spectroscopy reveals transient behavior, in which the TLS frequency fluctuates on the order of minutes. Thermal cycling above 10 K reverses these effects, restoring the TLS spectrum to its original state. Importantly, the intrinsic loss tangent of the LC oscillator remains unchanged before and after the application of the alternating bias. We propose that the disappearance of the steady-state spectrum is caused by nonequilibrium energy buildup from strain in the oxide film introduced by the pulsed voltage-bias sequence. Understanding this nonequilibrium energy could shed light on TLS fluctuations and reduce the calibration requirements for qubits.

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I. INTRODUCTION

Two-level systems (TLSs) are tunneling states in amorphous materials that contribute to energy loss and decoherence in superconducting and low-noise rf devices [1,2]. These effects are particularly detrimental to superconducting quantum bits (qubits), for which TLSs have been identified as a major source of decoherence. In 2005, Martinis *et al.* [3] discovered this connection, prompting extensive research by the qubit community into their origins and mitigation [1]. Subsequent studies have shown that

TLS-qubit interactions can persist for hours [4,5] and contribute to telegraphic noise and $1/f$ noise [6,7]. Beyond qubits, TLSs are a significant source of phase noise in microwave kinetic inductors used for astronomical photon detection [8–11], and they have been linked to charge noise in quantum dots [12].

One model that explains hourly TLS fluctuations invokes lower-frequency two-level systems referred to as two-level fluctuators (TLFs). These TLFs fluctuate because their energies are on the order of the temperature, and their physical proximity to resonant TLSs allows for mechanical coupling between the two [1,13]. TLFs are characterized by telegraph-like switching between two states, which can significantly shift the TLS frequency [13,14]. However, if many TLFs change simultaneously, the effect is no longer telegraphic. In superconducting devices used for dark matter detection and qubits, sudden bursts of phonons have been observed due to microfractures in the substrate resulting from stress released by adhesives such as the epoxy used to secure the devices to their packages [15–18]. Similarly, cosmic rays have been observed to randomize TLS frequencies [19]. While

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TLFs couple dispersively to resonant TLSs, they do not contribute to the total intrinsic loss [14,20]. TLSs that are resonant with qubits significantly decrease coherence times. Typically, qubits can be tuned away from these TLSs. However, random TLS fluctuations require constant retuning of the operating parameters. Understanding TLS time-dependent dynamics may shed light on material changes resulting from energy buildup and can lead to improved qubit calibration.

Recent studies have probed the effects of alternating voltage bias on device performance. Several studies have shown that they can manipulate the population [21,22] and the frequencies [23,24] of the TLS bath. Further studies have shown that applying a large alternating voltage bias changes the oxide barrier of Josephson junctions for superconducting qubits, offering a potential approach to improving device performance [25,26]. This alternating bias technique has been shown to accurately tune qubit frequencies by altering junction resistance. This change in resistance may be due to a nonreversible process in the junction caused by the migration and diffusion of Al^{3+} and O^{2-} ions within the barrier during the alternating processing. To amplify the effects of this biasing technique, previous experiments utilized a heated probing stage at temperatures up to 353 K [25–27].

This study investigates the impact of *in situ* cryogenic alternating bias stimulation (CABS) on TLS dynamics in an amorphous aluminum oxide ($\alpha\text{-Al}_2\text{O}_3$) film. We use an rf lumped-element LC oscillator in the strong coupling regime [28] to probe TLS dynamics. Similar platforms have been used to measure TLS dipole moments by adjusting the TLS energy within a large oxide barrier [29,30]. By applying an electric bias across the capacitor, we change the energy of TLSs within the bandwidth of our oscillator. The TLS-oscillator interaction is observed as a TLS-induced feature described by the standard tunneling model [31,32]. Using this model, we extract the electric dipole moment ($\vec{p}$) of the strongly coupled TLSs. After we apply CABS, TLSs no longer exhibit the characteristic steady-state spectral signatures observed upon initial cooldown [29,30]. Instead, analysis shows that the TLS-induced features are now irregularly distributed and change over time across the measurement bandwidth. In addition, loss tangent experiments conducted after each voltage and thermal treatment indicate that low-power loss does not change. Furthermore, after cycling to 10 K the TLS spectral signatures return to the pre-CABS state.

The remainder of this work is organized as follows. In Sec. II, we describe the design and operation of the experimental circuit, highlighting its sensitivity to TLSs embedded in trilayer capacitors. A review of our TLS spectroscopy and voltage-biasing procedures is provided. In Sec. III, we present TLS spectroscopy during different *in situ* CABS and thermal cycles. From these spectra, we extract dipole moments and densities using the standard

tunneling model. Loss tangent measurements after each voltage and thermal treatment are compared to understand changes in low-power loss and critical photon number. In Sec. IV, we discuss the implications of CABS on TLS dynamics and loss tangent measurements. Finally, we conclude by summarizing the key findings of TLS densities post-CABS and loss tangent measurements in Sec. V.

II. METHODS

A. Fabrication and device design

We use a lumped-element LC oscillator formed by a parallel-plate bias bridge capacitor [29,30,33] and an inductive element coupled to a feedline, as shown in Fig. 1. The device was fabricated on high-resistivity silicon using three lithographic steps. The bias bridge capacitor structure is a trilayer consisting of a 22 nm TiN film, a 49 nm amorphous aluminum oxide ($\alpha\text{-Al}_2\text{O}_3$), and a

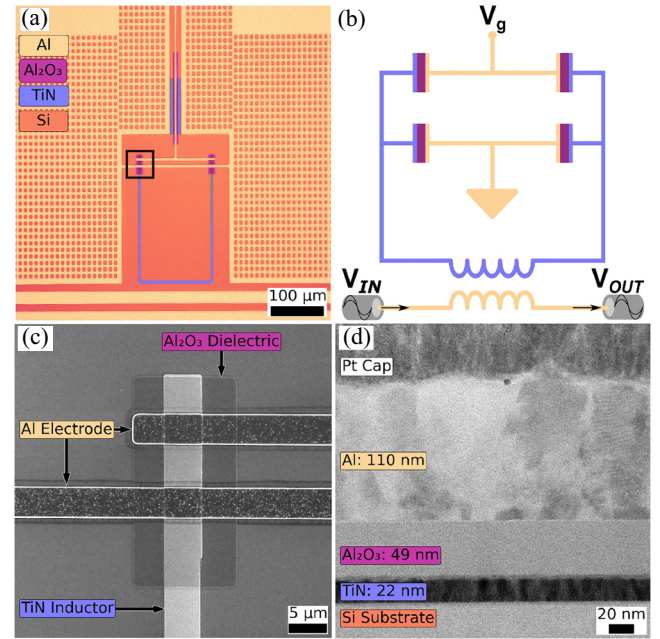


FIG. 1. (a) False-colored optical microscope image of the LC oscillator consisting of a Si substrate (orange), TiN (blue), Al_2O_3 (purple), and Al (yellow) layers. A voltage bias is applied along the thin, vertical Al channel. The TiN inductor is connected to four capacitive elements. The black box depicts two capacitors connected in a bias bridge. When biased, the capacitors have a constant dc field, $E_g = V_g/2d_o$, where $d_o = 49$ nm is the dielectric thickness. (b) A circuit diagram of the lumped-element resonator. The color coding depicts the same galvanic connections as in (a). (c) A scanning electron microscope image of the two capacitors highlighted in (a), acquired on a different oscillator of the same design. (d) A bright-field STEM image of the trilayer TiN- Al_2O_3 -Al capacitor cross section, capped with a Pt protective layer. The Pt cap was deposited to protect the capacitor during focused ion beam cross-section isolation and was not present during device TLS characterization.

~ 110 nm Al capping layer [see cross-section image in Fig. 1(d)]. The TiN layer was deposited using plasma-enhanced atomic layer deposition (ALD). The oxide layer was deposited using thermal ALD, while the Al layer was added via electron beam evaporation. It should be noted that the ALD-grown oxide barrier in this study differs from the commonly employed oxide growth method for Josephson junction-based qubit fabrication, which is a diffusion-limited oxidation of an aluminum layer [34]. ALD forms oxide layers by building up the oxide sequentially, whereas electron beam evaporation produces oxides in a single deposition step. While we could not exactly duplicate the oxidation conditions in the JJ, we chose to use ALD because of the uniformity of the amorphous oxide [35]. Having a uniform oxide allows us to isolate a single TLS-forming environment, potentially simplifying our analysis.

The inductive element is a single U-shaped inductor made of TiN, which couples to a feedline with a coupling quality factor $Q_c \approx 6500$. Due to the TiN's higher kinetic inductance ($189 \text{ pH}/\square \pm 3 \text{ pH}/\square$) compared with aluminum, the inductive element is shorter than an equivalent Al inductor. An equivalent Al inductor would need to be longer and more meandered, which would increase parasitic capacitance and reduce the participation of the trilayer capacitors. The Al layer also forms the ground plane, upper capacitive electrodes, and bonding pads. To mitigate flux noise, the ground plane was perforated. The trilayer design minimizes electromagnetic coupling to stray noise sources and concentrates the electric field within the small oxide volume ($5.3 \text{ } \mu\text{m} \times 5.3 \text{ } \mu\text{m} \times 49 \text{ nm}$) [see Figs. 1(c) and 1(d)]. This concentrated electric field allows us to strongly couple to the TLSs and the majority of loss originates from TLSs within the oxide barrier [1]. Additional fabrication details are provided in Appendix A.

B. Device operation

Initial measurements of the oscillator were performed in the first dilution refrigerator, which we designate as the “control” condition. After approximately 5 months, the device was transferred to a second dilution refrigerator before further experiments were conducted. We refer to this period as the “control-aged” stage to account for any potential aging effects on device parameters. Aside from relocating the device, all other experimental conditions and setup parameters in the second dilution refrigerator remained unchanged for the remainder of the study.

The bias line (V_g) is placed in a virtual ground between the capacitors, minimizing coupling to the rf mode [Figs. 1(a) and 1(b)]. The voltage bias generates a uniform electric field $E_g = V_g/(2d_0)$ across the bias bridge, where d_0 is the dielectric thickness. The capacitors were designed with a small volume to achieve strong coupling between TLSs and the oscillator [28].

The rms electric field in the capacitors is calculated as $E_{\text{rms}} = \sqrt{\hbar\omega_c/2\epsilon V_T}$, where V_T is the total capacitor volume and ω_c is the oscillator frequency [28]. The dielectric constant of the oxide is $\epsilon = \epsilon_r\epsilon_0$, with $\epsilon_r \approx 10$. The capacitor volume V_T in this study is $5.6 \text{ } \mu\text{m}^3$, significantly smaller than the $78 \text{ } \mu\text{m}^3$ reported in [29], resulting in an E_{rms} of 53 V/m , approximately $2.5\times$ stronger than previous work.

III. RESULTS

A. TLS spectroscopy

To quantify TLS-oscillator interactions, we measured the transmission spectrum (S_{21}), as shown in Fig. 2. In some spectra, peaks in the Lorentzian profile indicate interference caused by TLS coupling to the oscillator [Fig. 2(a)] [13,28–30,36]. To isolate TLS-induced features, we averaged out the oscillator's Lorentzian profile and time-varying components along the voltage axis (see Appendix C).

The applied electric field bias E_g modifies the TLS energy according to Eq. (1)

$$\epsilon = (\Delta_0^2 + (\Delta - 2\vec{p} \cdot \vec{E}_g)^2)^{1/2}, \quad (1)$$

where Δ_0 represents the tunneling barrier height, Δ is the TLS asymmetry energy, and $\vec{p}$ is the TLS dipole moment [29–32].

Figure 2(b) shows a measurement of the oscillator spectrum as the TLS energy is tuned via the applied voltage bias V_g . Each TLS spectrum required approximately 50 hours to complete, with measurements performed with an intermediate frequency bandwidth of 1 Hz to ensure sufficient averaging of TLS-induced features. These measurements, conducted prior to any treatment steps and in the second dilution refrigerator (see Sec. II A), serve as the control for the remainder of this study. All measurements, including those depicted in Fig. 2, were conducted at the single-photon level [37].

To analyze the TLS spectra over a large voltage range, image processing and a machine learning algorithm were employed to identify individual hyperbolic curves. To optimize the fit to the TLS spectrum, we first implemented a denoising step and a gradient edge filtering scheme to enhance the definition of TLS-induced features. The machine learning algorithm then identified TLS-induced hyperbolic features, which were fit using Eq. (1). Dipole densities were calculated from the extracted TLS dipole moment statistics and compared across different device treatments. Additional details on the TLS fitting procedure are provided in Appendix C.

We extracted TLS-induced hyperbolas from the control with an average dipole moment of $0.49 \text{ e}\text{\AA}$ [Fig. 3(a)] and a TLS density of $75 \pm 10 \text{ TLS}/(\mu\text{m}^3 \text{ GHz})$. After the device was aged for 5 months, the average TLS dipole moment

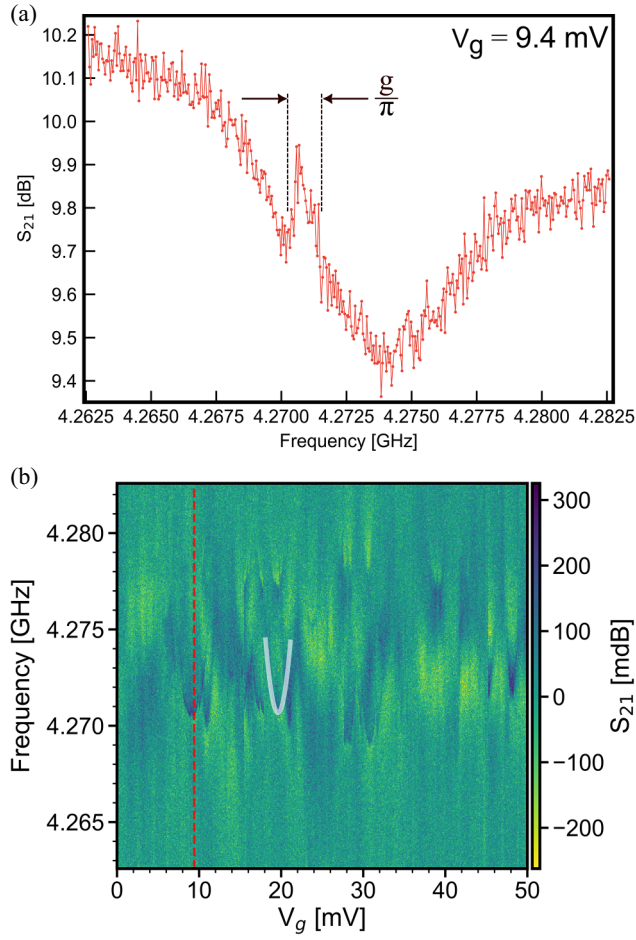


FIG. 2. (a) Transmission (S_{21}) of an oscillator interacting with a TLS at $V_g = 9.4$ mV. The width of the TLS-induced features is highlighted with black dashed lines, showing the outcome of the oscillator's coupling to the TLS. The TLS-oscillator coupling strength g/π is calculated to be 1.3 MHz, which matches the width of the TLS-induced feature. (b) Transmission (S_{21}) as a function of applied voltage (V_g) and oscillator frequency. The TLS-oscillator interactions appear as hyperbolic shapes following Eq. (1). An example is shown with the white curve corresponding to an extracted dipole moment $p_z = 0.24$ eÅ. The individual Lorentzian spectrum shown in (a) is a line cut along the red dashed line. Data processing for this figure is described in Appendix C.

dropped to 0.31 ± 0.02 eÅ, but the density was similar, 93 ± 24 TLS/ $(\mu\text{m}^3 \text{ GHz})$ [see Fig. 3(b)]. Statistical analysis of the control experiment confirmed that our results are consistent with previous studies on TLS dipole moments and densities in oxide films [29,30].

The TLS-oscillator coupling strength, $g = p_z \sqrt{\omega_c/2\epsilon\hbar V_T}$ can be calculated using the extracted dipole moment [3,28]. Here, p_z is the projected dipole moment along the applied E_g direction. Using the average dipole moment of the control $p_z = 0.5$ eÅ, the TLS coupling strength is calculated as $g/\pi = 1.3$ MHz, which matches the width in Fig. 2(a).

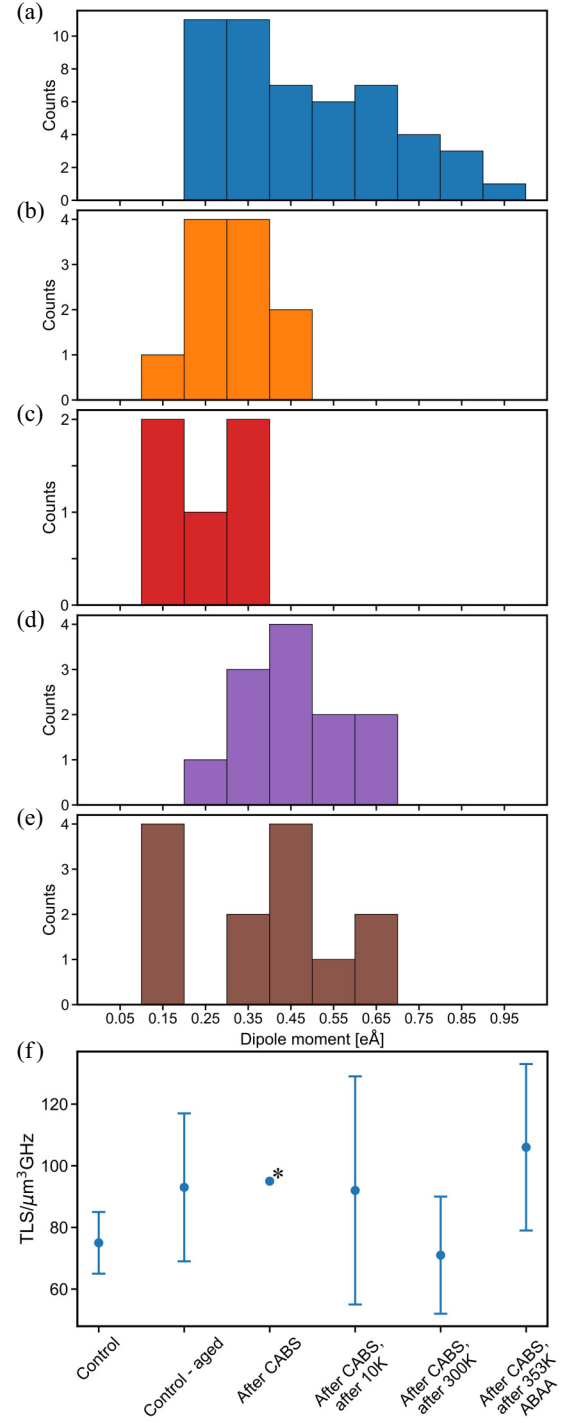


FIG. 3. Extracted dipole moments for TLS spectra collected after different treatments. The histograms represent the following: (a) control (blue, initial measurement), (b) control-aged, (orange, after aging 5 months), (c) 10 K temperature cycle after CABS (red), (d) 300 K temperature cycle after CABS (purple), and (e) after a 353 K alternating bias assisted annealing treatment (brown). We do not present any data on electric dipole moments directly after CABS because there were no TLS-induced hyperbolas found in this scan. (f) The extracted TLS densities for the different processing steps. *Calculated value of TLS density from the main paper after CABS treatment.

B. Alternating bias treatments

To investigate the effects of alternating bias on TLSs at cryogenic temperatures, we applied a time-varying bias to the voltage gate following the procedures described in [25,26]. Alternating pulses of ± 10 V were applied for 10 s each, repeated 2800 times over 30 h. Each pulse was separated by a 10 s dwell time at 0 V, creating an electric field of $E_g = \pm 100$ MV/m across the oxide [Fig. 1(d)].

Following the treatment, TLS spectroscopy was conducted over a voltage range of 0–50 mV with 100 μ V steps, taking a total of 50 h. The results, shown in Fig. 4(a), do not present any TLS-induced hyperbolas. However, Fig. 4(a) reveals sharp, unidentified features in the TLS spectra. Figures 4(b)–4(e) are line cuts of Fig. 4(a) at specific bias points.

Closer inspection of the resonator transmission reveals TLS-induced features with magnitudes similar to those observed in the control spectra (Fig. 2). The estimated coupling strength (g/π) associated with these features is

approximately 1 MHz. However, these TLS-induced features are highly transient, lasting less than 5 min. Since each scan takes 6 min this measurement only provides an upper bound on the TLS frequency jitter. The next curve typically does not capture a TLS at the same frequency.

Despite Fig. 4 not presenting hyperbolas, we were able to extract approximate TLS parameters using the TLS-induced feature width as we did in Fig. 2(a) (see Table I). In total, 122 individual TLS-induced features were found within a 12 MHz bandwidth of the oscillator, out of 500 different applied bias points. Electric dipole moments could not be fit to these features because full hyperbolas were not observed. Instead, we compared the CABS sample and the control sample using the ratio of TLS-induced features to total bias points. One interpretation of the CABS spectrum is that at each instant of a TLS-induced feature, a different resolvable TLS hyperbola, statistically independent from others, would be observed if the measurement were instantaneous. With this assumption, we

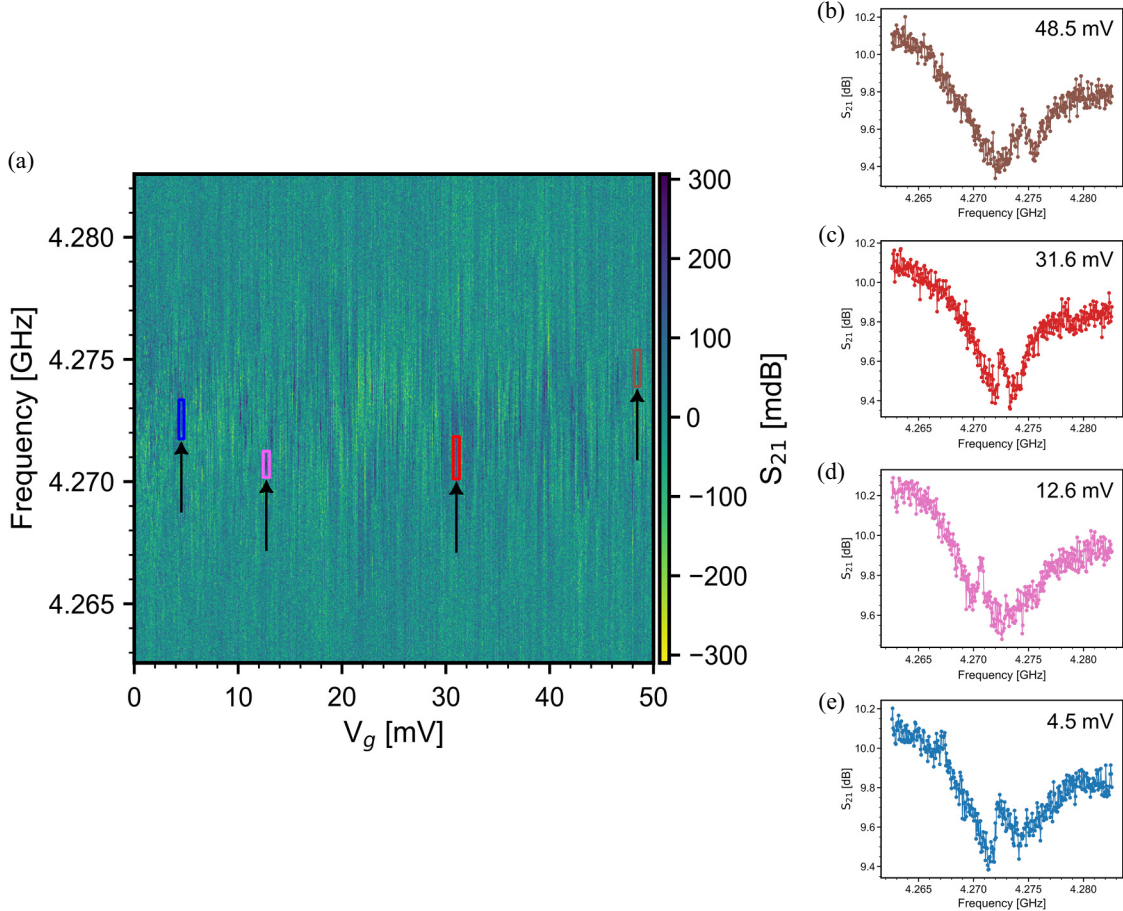


FIG. 4. (a) Transmission (S_{21}) versus applied voltage (V_g) and oscillator frequency spectrum after the CABS treatment reveals sharp features during the voltage scan. Post-CABS treatment reveals no TLS-induced hyperbolas. Instead, the TLS spectrum appears to exhibit frequency shifts, with transient TLS-induced features lasting less than 5 min. Data processing for this figure is described in Appendix C. (b)–(e) Individual transmission spectra at the points indicated in (a). TLS-induced feature with a width of approximately 1 MHz can be observed.

measured 122 TLS in 500 curves, or a probability of 0.24 TLS per transmission spectrum. Before CABS, the measurements were not statistically independent. A typical TLS hyperbola has a width of 0.75 mV, and we measured approximately 50 TLS over 200 mV. Therefore, at a random bias there are 0.19 TLS per transmission spectrum.

Although the TLS landscape is significantly altered by the CABS procedure, the density of TLS-induced features throughout the bias sweep seems to be comparable. Assuming the same dipole moment distribution as the control sample, the TLS density of the CABS sample would be 95 TLS/(μm^3 GHz), similar to that of the control sample [30, Eq. (A14)].

C. Thermal cycling

Following the CABS treatment, we investigated the effects of thermal cycling to 10 K. The spectroscopy is shown in Fig. 5. After temperature cycling we measured a density of 92 ± 37 TLS/(μm^3 GHz), [Fig. 3(c)].

Similarly, after cycling to 300 K (room temperature), the TLS density and average electric dipole moment had values similar to those observed in the pre-CABS state [see Table I and Fig. 3(d)].

We next explored the influence of TLS density using a noncryogenic alternating voltage-bias treatment [25,26], performed on a probe station for 30 h. During this test, the platform supporting the device was heated to 353 K for the entire duration. A ± 10 V alternating bias was applied using 1 s pulses to minimize the risk of dielectric breakdown. TLS spectroscopy conducted after this treatment revealed a density of 106 ± 27 TLS/(μm^3 GHz) (see Appendix D).

Figure 3(e) presents a summary of the dipole moment distributions, and Table I lists the densities. The average electric dipole moment after the 353 K alternating bias treatment was 0.38 ± 0.05 eÅ. This electric dipole moment was similar to the average dipole moment in the initial control and after the 300 K cycle measurement. No

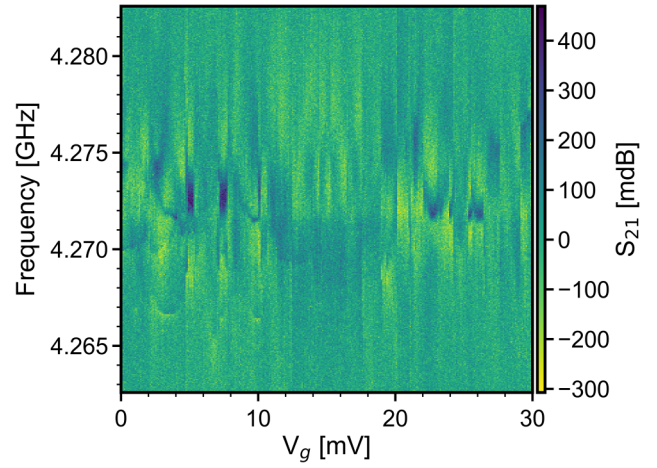


FIG. 5. Measured transmission spectrum (S_{21}) after thermal cycling to 10 K. TLS-induced hyperbolas can be seen in this spectrum. Data processing for this figure is described in Appendix C.

matter the treatment we applied, all the densities were very similar.

D. TLS low-power saturation

To further understand the effects of CABS on TLSs, we studied the power dependence of TLS loss [9]. Figure 6 shows the loss tangent, $\tan \delta$, as a function of the microwave energy injected into the resonator. Measurements were performed at each stage of sample processing: control, CABS, 10 K thermal cycle, and 353 K alternating bias treatment [25,26]. The control measurement was conducted on the same sample but in a different cryostat.

The loss tangent curve was fitted using Eq. (2)

$$\tan \delta = \frac{\tan \delta_{\text{TLS}}^0}{\sqrt{1 + \frac{n}{n_c}}} + \tan \delta_E, \quad (2)$$

TABLE I. Average TLS electric dipole moments (p_z), TLS densities, low-power loss tangent values, and critical photon number for different *in situ* and thermal treatments. Two loss tangents are provided, one measured from power saturation and the other calculated from TLS density using Eq. (A13) [30]. Errors represent one standard deviation from the mean.

Treatment	mean p_z (eÅ)	TLS density ($\frac{\text{TLS}}{\mu\text{m}^3 \text{ GHz}}$)	$\tan \delta_{\text{TLS}}^0 (\times 10^{-3})$	$\tan \delta_{\text{TLS}}^{\text{calc}} (\times 10^{-3})$	$n_c (\times 10^{-3})$
Control	0.49 ± 0.03	75 ± 10	1.89 ± 0.03	0.21 ± 0.03	...
Control-aged (Aged 5 months)	0.31 ± 0.02	93 ± 24	^a	0.12 ± 0.03	^a
CABS	...	^b 95	2.00 ± 0.03	...	22 ± 2
10 K thermal cycle	0.24 ± 0.04	92 ± 37	2.00 ± 0.05	0.07 ± 0.03	23 ± 2
300 K thermal cycle	0.45 ± 0.04	71 ± 19	^a	0.14 ± 0.04	^a
Alternating V_g at 353 K	0.38 ± 0.05	106 ± 27	1.64 ± 0.03	0.17 ± 0.04	17 ± 2

^aNot measured.

^bCalculated from the analysis of TLS-induced features in Fig. 4.

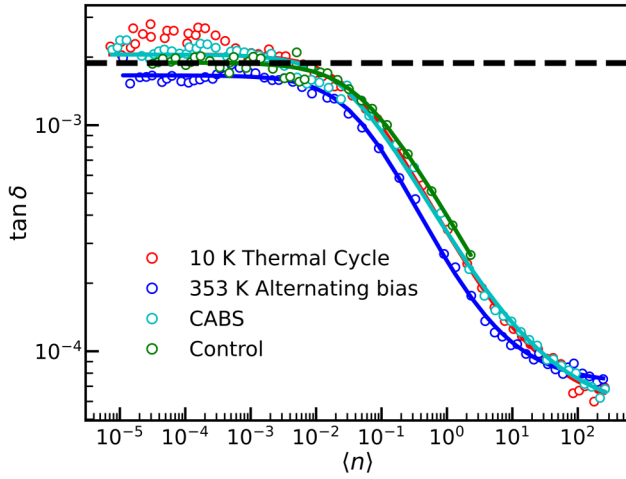


FIG. 6. Loss tangent ($\tan \delta$) versus average photon number ($\langle n \rangle$) under various experimental conditions. Loss tangent curves are shown for the control (green circles), after CABS treatment (teal circles), after 10 K thermal cycling (red circles), and after alternating voltage-biasing annealing at 353 K (blue circles). All measurements were conducted at a base temperature of 10 mK. Solid lines represent fits to the loss tangent data using Eq. (2), with colors corresponding to each treatment step. The black dashed line indicates the average intrinsic TLS loss ($\tan \delta_{\text{TLS}}^0 = 1.88 \times 10^{-3}$), which provides a baseline for comparison.

where n and n_c are the average photon number and critical photon number, respectively, and $\tan \delta_E$ is the power-independent contribution to the loss tangent. From the standard tunneling model, the low-power loss tangent due to TLSs is $\tan \delta_{\text{TLS}}^0 = \pi P_0 |\vec{p}|^2 / 3\epsilon$, where ϵ is the dielectric permittivity of the oxide layer and P_0 is the TLS density [1,38]. The average $\tan \delta_{\text{TLS}}^0$ was calculated as $(1.88 \pm 0.07) \times 10^{-3}$ (black dashed line in Fig. 6) and was consistent across all processing steps (see Table I). This value aligns with previous measurements for alumina, where $\alpha\text{-Al}_2\text{O}_3$ exhibited loss tangents of 1.50×10^{-3} and $\alpha\text{-AlO}_x$ exhibited 0.98×10^{-3} [30].

IV. DISCUSSION

The initial TLS spectroscopy revealed TLS-induced hyperbolas (Fig. 2), consistent with previous reports [13, 28–30]. These features persisted for several hours, in agreement with observations in superconducting qubits [4,5]. Based on the density of TLS-induced hyperbolas in Fig. 2(b), the contribution of strongly coupled TLSs to the loss tangent is calculated to be $\tan \delta_{\text{TLS}}^0 = 0.12 \times 10^{-3}$, which is an order of magnitude lower than the fitted value in Fig. 6. The calculation of $\tan \delta_{\text{TLS}}^0$ was done using Eq. (A13) in Ref. [30]. This calculated result for $\tan \delta_{\text{TLS}}^0$ is on the order of the error margin of the fit (Fig. 6), indicating that visible strongly coupled TLSs are a minor correction to the overall background loss. In addition, following

CABS and other treatments, the $\tan \delta_{\text{TLS}}^0$ fits (Fig. 6) converged to the same value, suggesting consistent behavior across the TLS population.

Our calculated TLS ac coupling $g/\pi = 1.3$ MHz agrees with the measured TLS-induced features width seen in Fig. 2(a) for the rms driven electric field of $E_{\text{rms}} = 53$ V/m in our capacitors. The driven field is calculated at the single-photon regime. Assuming that the n_c extracted from Fig. 6 corresponds to strongly coupled TLSs, the TLS relaxation rate T_1^{TLS} is approximately $1 \mu\text{s}$ when we use our measured $n_c = 21 \times 10^{-3}$, $p_z = 0.5$ eÅ, and $2\pi g$. This T_1^{TLS} agrees with previous studies of strongly coupled TLSs [33,39–41]. From our measured TLS ac coupling and device statistics, we show consistency with other TLS measurements of amorphous oxides.

Following this initial measurement, we applied CABS to the dielectric film (see the Results section for details). The application of CABS follows a protocol similar to that in Refs. [25,26]. After CABS, the TLS-induced hyperbolas were absent and could not be fit to Eq. (1). Instead, the spectroscopy displayed randomly distributed TLS-induced features [see Figs. 4(b)–4(e)]. The calculated ratio of TLS-induced features to total bias points was within a factor of 1.3, as determined by individually counting TLSs with coupling strengths (g/π) and TLS-induced feature depths comparable to those shown in Fig. 2(a) (see Results). If each TLS-induced feature corresponds to a distinct TLS, the estimated density is approximately 95 TLS/(μm^3 GHz). These results indicate that although hyperbolic features are absent from the TLS spectrum after CABS, the overall TLS distribution likely remains unchanged.

Strongly coupled TLSs reappeared after thermal cycling to 10 K and subsequent cooling to 10 mK (Fig. 5), with extracted dipole moments ranging from 0.5 to 0.35 eÅ. These TLSs once again fluctuate on the order of hours. The 10 K thermal cycling experiment exhibited a TLS density comparable to the values observed in the control experiment. These results are consistent with previous experimental observations, such as the breakdown of the TLS model above 15 K in sound speed measurements for $\alpha\text{-GeO}_2$ and $\alpha\text{-B}_2\text{O}_3$ [42], which indicated temperature-dependent changes in TLS behavior. There has also been a recent study by Zanuz *et al.* showing that TLS defect modes in qubits rearranged after thermal cycling to room temperature [43]. Our findings complement these prior works by demonstrating the reversible nature of strongly coupled TLSs in oxide films at temperatures above 10 K. The reappearance of TLSs after thermal cycling suggests that the TLS ensemble undergoes structural or energetic reorganization at elevated temperatures. These results indicate a dielectric transition associated with overcoming the barrier height.

Furthermore, the TLS-induced features after CABS were highly transient. Since each scan took 6 min, this

measurement provides an upper bound on the TLS frequency jitter of approximately 3 mHz. Typically, TLSs fluctuate on the order of hours, but after CABS their fluctuations are approximately 100 times faster. The observed spectral fluctuations also do not show telegraphic or diffusive drift of TLS-induced features, in contrast to previous superconducting qubit measurements [4,5]. Due to these dynamics, we postulate that the TLF bath coupling to the TLS bath is in a nonequilibrium state that evolves between measurements. Previous results demonstrating TLS scrambling include cosmic-ray impacts [19] and hole burning in the TLS density of states [44,45]. We believe that in our experiments, energy storage in the dielectric is responsible for the TLS transient behavior.

The transient TLS behavior suggests that the system entered a metastable state. The application of bias likely injected nonequilibrium energy, which is stored in the device through some mechanism. The intermittent release of that energy causes TLSs to shift in frequency. This would explain the constant $\tan \delta_{\text{TLS}}^0$ since it would not directly change the density of TLSs.

One guess as to the energy storage mechanism is the buildup of stress in the structural configuration of the amorphous material. Alternating bias treatments have been shown to alter the atomic profile of the dielectric material [25], potentially inducing piezo-tunneling effects in materials such as α - Al_2O_3 . It is also interesting to note that the field strength used for CABS is of the same order of magnitude as the parasitic electric fields generated by trapped charges in ALD-grown α - Al_2O_3 junctions [46]. Stress-induced relaxation events, likely phonon bursts into the dielectric, could release this energy randomly after CABS is applied, and gradually settle toward some equilibrium [15–18]. Mechanical strain has been shown to mediate the TLS-TLF interactions [14]; and phonon bursts resulting from cosmic rays have been shown to scramble TLSs [19]. Therefore, it is conceivable that the nonequilibrium energy buildup is stressing the atomic lattice and releasing periodic phonon bursts that are scrambling the TLS frequencies.

V. CONCLUSION

This study introduced CABS as an *in situ* treatment that modifies the defect environment in a dielectric film. Building on recent findings that alternating voltage biasing may indicate a structural change within qubits at 353 K [25], we applied CABS for 30 h and observed that TLSs are no longer stable in frequency. What remains in the spectrum after CABS are random TLS-induced features that fluctuate on the order of minutes.

Our analysis following CABS processing suggests that the TLS density remained constant. Interestingly, when the sample was thermally cycled above 10 K, we observed

TLS frequency stability, demonstrating the reversible nature of Al_2O_3 films.

We propose that CABS causes a nonequilibrium energy buildup in the dielectric. The release of this energy leads to TLS fluctuations on the order of minutes, about 100 times faster than pre-CABS levels. Similar energy buildup and release have been observed in phonon-only events from microfractures caused by device stress [15–18]. Our method of repeatedly injecting nonequilibrium energy sheds light on the origins of TLS fluctuations, which is essential for developing fault-tolerant and efficient qubit architectures.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX A: FABRICATION DETAILS

The devices are fabricated on high-resistivity intrinsic Si (100) wafers. Prior to fabrication, the wafers were cleaned with a preliminary piranha dip, followed by a standard RCA wafer cleaning process [47], and then a final dilute HF dip (49% HF diluted 50:1 with de-ionized water) to remove the native oxide. The oscillator's base layer, serving as the inductor, consists of a 22 nm TiN film deposited by plasma-enhanced atomic layer deposition (ALD) with the substrate chuck maintained at 275 °C, using tetrakis(dimethylamido)titanium (TDMAT) at 75 °C and plasma N_2 as precursors. The kinetic inductance is calculated to be $189 \text{ pH}/\square \pm 3 \text{ pH}/\square$, where the uncertainty is estimated from the frequency spread observed across devices. The base layer is patterned using photolithography with a Nikon i-line stepper and etched with a BCl_3/Cl_2 plasma.

A 49 nm Al_2O_3 dielectric layer was deposited by thermal ALD at a substrate temperature of 250 °C, using room-temperature precursors trimethylaluminum (TMA) and H_2O . This layer is patterned by photolithography and etched with a wet phosphoric acid etch (Transetch-N). The thickness uncertainty of the ALD-deposited layers is ± 4 nm, with an assumed film uniformity variation of $\sim 1\%$.

The device's top metallization, comprising the ground plane, bias lines, bond pads, and the capacitor's upper electrode, is defined using image-reversal photoresist and the i-line stepper with a flood exposure. A 150 nm aluminum layer is deposited by e-beam evaporation and lifted off to complete the structure.

APPENDIX B: FRIDGE AND MEASUREMENT LAYOUT

To measure transmission (S_{21}), we routed a microwave signal through a dilution refrigerator equipped with multiple stages of attenuation totaling 60 dB and a 10 GHz low-pass filter (see Fig. 7). The attenuation stages were used to minimize thermal noise. The device sample, measuring 8 mm $\times$ 8 mm, was mounted inside a gold-plated copper (Cu) enclosure to provide thermal and electrical stability. Dc bias and microwave signals were delivered to the chip via sub-Miniature push-on (SMP) connectors and wire bonds, with GE varnish a common commercial varnish intended for cryogenic thermometry supplied

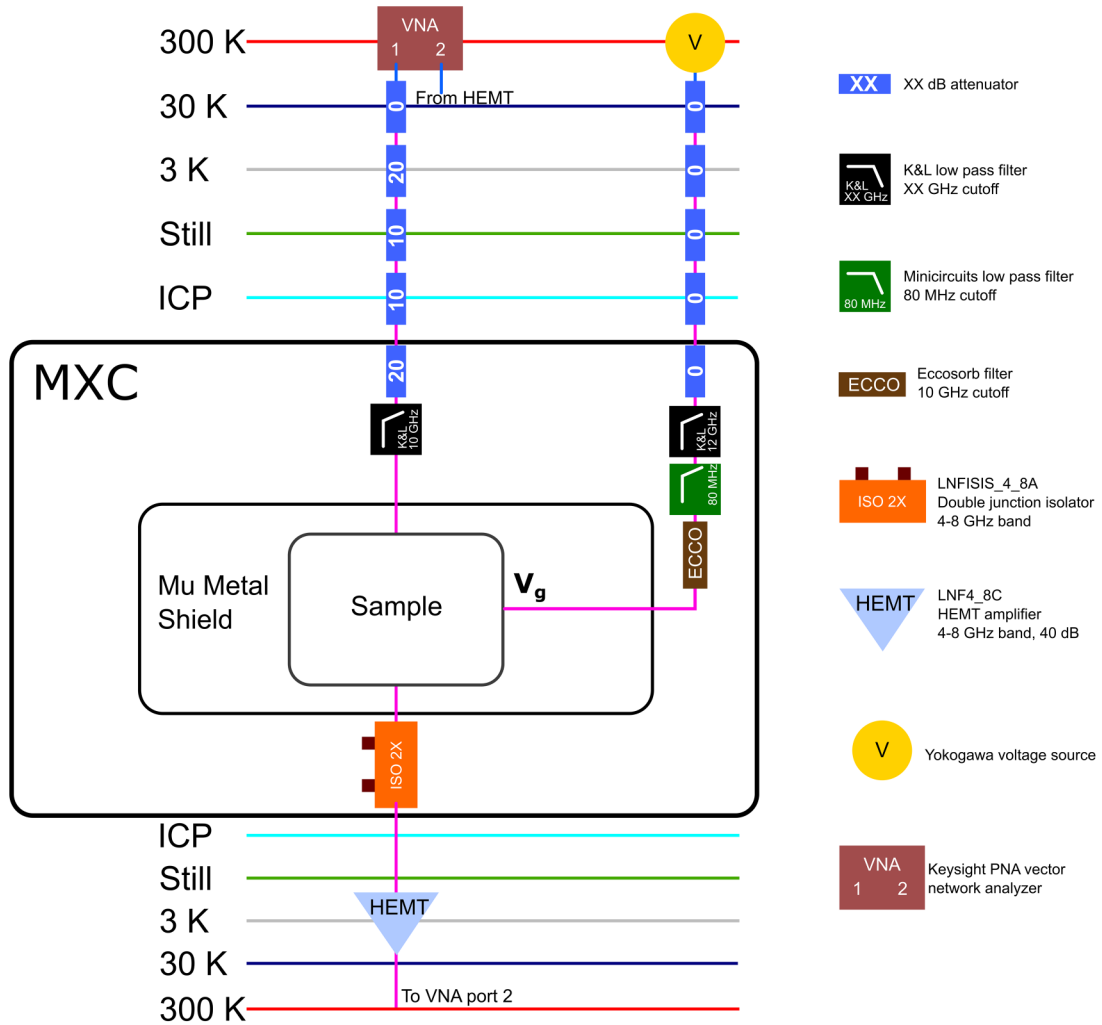


FIG. 7. The wiring diagram illustrates the experimental setup for the oscillator device, highlighting key components and their roles in ensuring optimal performance. The microwave signal path includes filters, isolators, and other rf components, which are labeled on the right-hand side of the diagram. Attenuation values (indicated by blue squares) are specified for each temperature stage, with a total attenuation of 60 dB. The sample is on a paddle mounted on the mixing chamber (MXC) stage, which is cooled to a base temperature of 10 mK using a dilution refrigerator. Surrounding the MXC stage is a magnetic shield made of mu-metal, designed to attenuate stray magnetic fields that could interfere with the device's operation. Voltage dc biasing (V_g) is applied through a coaxial line with no attenuation.

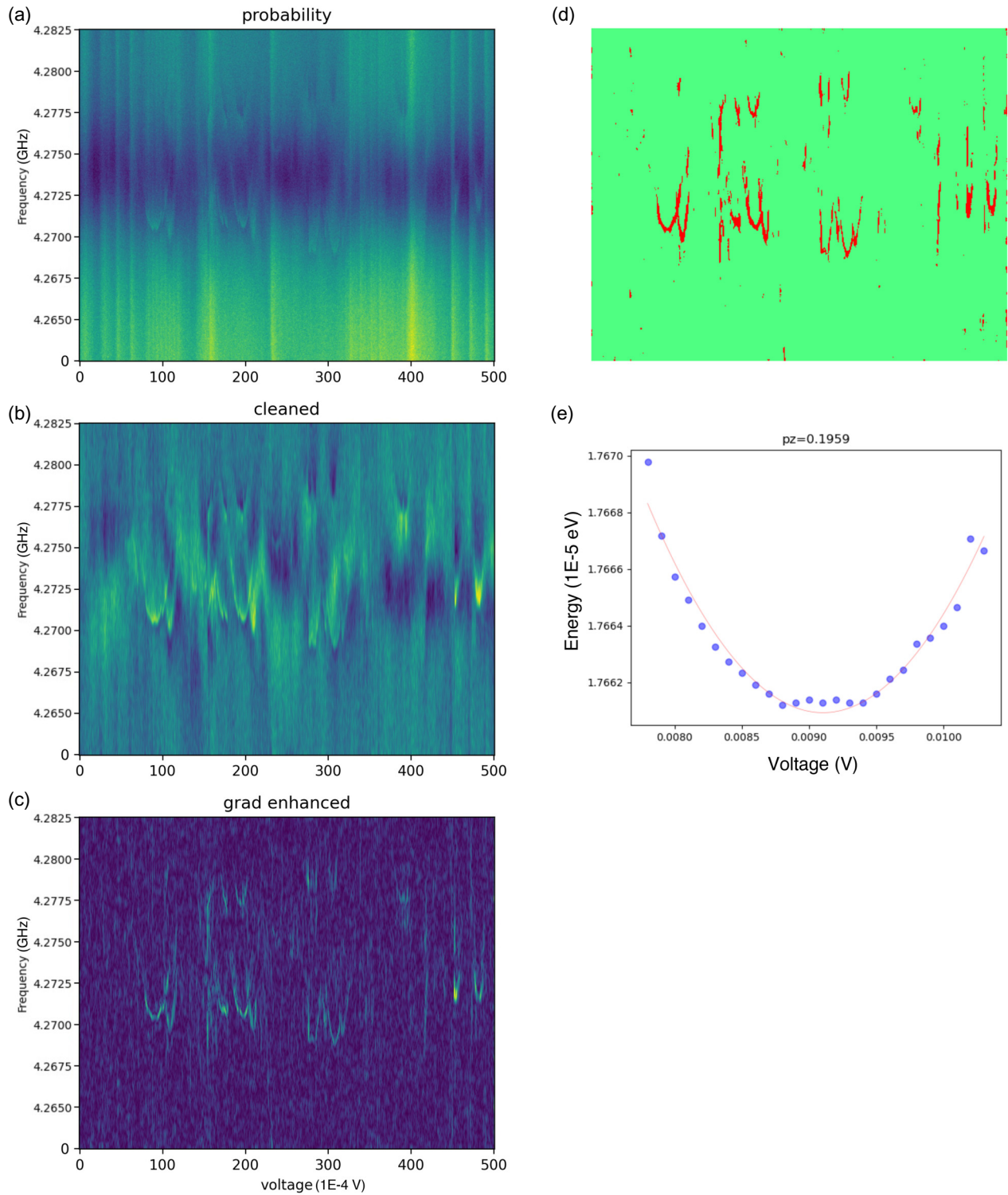


FIG. 8. (a) Unprocessed experimental data. (b) Cleaned data, with fluctuations in time and in frequency removed as described in Appendix C. (c) Data after gradient enhancement. (d) Output of the ML model. (e) Example of a fitted hyperbola, where the solid red line denotes the fit and the blue dots are taken from one hyperbola in (d).

by Lakeshore Cryotronics (VGE-7031) used to secure the device to the Cu housing.

Further down the signal chain, the microwave signal passed through two blocks of cryogenic isolators operating

in the 4–8 GHz range to suppress back reflections and noise. A cryogenic amplifier (high electron mobility transistor, HEMT) amplified the signal by approximately 35 dB, ensuring sufficient signal strength for accurate

measurement. The device was mounted on a paddle attached to the MXC stage of a Bluefors XLD-1000 dilution refrigerator, as shown in Fig. 7. The dilution refrigerator was operated at a base temperature of 10 mK. Surrounding the paddle at the MXC was a magnetic shield made from mu-metal, which effectively blocked stray magnetic fields that could interfere with the device's operation.

To tune the energy of TLSs, we employed a voltage-bias line with no attenuation. The bias line included two stages of low-pass filtering: an Eccosorb filter to suppress high-frequency noise and a Minicircuits 80 MHz low-pass filter for additional noise reduction. The dc voltage bias was supplied using a Yokogawa GS200, providing precise and stable control over the applied voltage.

APPENDIX C: TLS FITTING DETAILS AND MACHINE LEARNING ALGORITHMS

Transmission spectra (S_{21}) plots were processed using several steps before fitting the TLS energy vs applied electric bias hyperbolas, Eq. (1) from the main paper, to features in those plots. These steps are summarized below and the results are shown in Fig. 8.

The Lorentzian around the resonator frequency in the S_{21} spectra was removed by subtracting the S_{21} value at each frequency averaged over bias: $\tilde{P}(f, V) = P(f, V) - \bar{P}_{\text{bias}}(f)$. Similarly, the baseline shift over time was removed by subtracting the frequency-wise average: $\tilde{P}'(f, V) = \tilde{P}(f, V) - \bar{P}_{\text{frequency}}(V)$. Then we applied a 1D Gaussian filter from the scipy python package and the result of this step is shown in Fig. 8(b). The transmission spectra in the main paper and Fig. 9 have only undergone the two averaging steps to visually depict changes in the TLS-induced hyperbolas.

We next calculate the magnitude of the gradient of $\tilde{P}'(n)$ with respect to the image array coordinates (i, j) , where i and j refer to bias and frequency:

$$|\nabla \tilde{P}'(i, j)| = \sqrt{\left(\frac{\partial \tilde{P}'(i, j)}{\partial i}\right)^2 + \left(\frac{\partial \tilde{P}'(i, j)}{\partial j}\right)^2}.$$

By applying this technique, we enhanced the contrast of the TLS hyperbola traces in the cleaned image [Fig. 8(c)].

The resulting image was processed using the Trainable Weka Segmentation plugin for the Fiji image processing package based on the ImageJ2 software. The machine learning (ML) model was trained by manually selecting the portions of the image that correspond to TLS hyperbolas and those that do not, by hand. The algorithm used in the Trainable Weka Segmentation plugin is the fast random forest classifier. The resulting classified image is shown in Fig. 8(d). With this image we select a rectangular region for each hyperbola by hand, and pixels in those regions are

fit to Eq. (1) from the main paper in order to extract the TLS parameters: Δ_0 , Δ , and p_z [Fig. 8(e)]. The TLS densities were extracted from dipole moment histograms using the method detailed in Ref. [30].

TLS hyperbolas are only counted if Δ_0 is within the bandwidth, therefore the primary source of uncertainty in the TLS density is due to the number of TLS with Δ_0 within the bandwidth sampled. We divided the bandwidth into bins and calculated the standard deviation of the populations of TLS with Δ_0 in those bins. From this, we estimated the uncertainty in the total number of TLS with Δ_0 in the bandwidth by dividing that bin-wise standard deviation by the square root of the number of bins in the bandwidth.

APPENDIX D: POST-ROOM-TEMPERATURE THERMAL CYCLE AND ALTERNATING VOLTAGE BIAS

Figure 9 presents the results obtained after the device underwent thermal cycling to 300 K (room temperature) and was subjected to an alternating bias treatment, as described in the Methods section and detailed in [25]. During the experiment, the device was mounted on a voltage probe station, where an alternating bias treatment, as described in Methods and detailed in [25], was applied at 353 K for 30 h. After cooling the device back down to

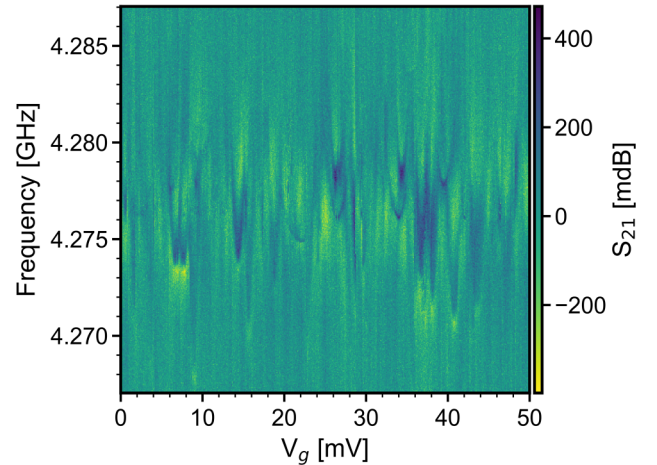


FIG. 9. Transmission (S_{21}) versus applied voltage (V_g) and oscillator frequency was measured after alternating voltage-bias treatment. The treatment was conducted with a probe station and a hot plate set at 353 K, following the process described in Refs. [25,26], but with ± 10 V bias pulses and a 1 s dwell time between pulses for a total duration of 30 h. After treatment, the device was mounted in a dilution refrigerator, and the transmission spectrum was measured at a base temperature of 10 mK. The extracted dipole moments ranged from 0.15 to 0.65 eÅ, with a dipole density of 106 ± 27 TLS/(μm^3 GHz). Data processing for this figure is described in Appendix C.

10 mK, TLSs with dipole moments of 0.15–0.65 eÅ and a density of 106 ± 27 TLS/(μm^3 GHz) were measured.

These results indicate that the TLS energy landscape remained consistent with measurements taken after the 10 K thermal cycle (see Table I and Fig. 3 in the main paper). Specifically, the alternating bias treatment did not significantly alter TLS properties, as evidenced by the unchanged loss tangent value ($\tan \delta_{\text{TLS}}^0 = 1.64 \times 10^{-3}$). This suggests that the alternating bias treatment preserves the TLS landscape under the tested conditions and does not significantly affect the strongly coupled TLSs.

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