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Quantum paraelectricity probed by superconducting resonators

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Superconducting coplanar waveguide (CPW) resonators are powerful and versatile tools used in areas ranging from radiation detection to circuit quantum electrodynamics. Their potential for low intrinsic losses makes them attractive as sensitive probes of electronic properties of bulk materials and thin films. Here we use superconducting MoRe CPW resonators to investigate the high-frequency (up to 0.3 GHz) and low-temperature (down to 3.5 K) permittivity of SrTiO₃, a nonlinear dielectric on the verge of a ferroelectric transition (quantum paraelectricity). We perform a quantitative analysis of its dielectric properties as a function of external dc bias (up to ± 15 V), rf power, and mode number and discuss our results within the framework of the most recent theoretical models. We also discuss the origin of a fatigue effect that reduces the tunability of the dielectric constant of SrTiO₃, which we relate to the presence of oxygen vacancies.

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I. INTRODUCTION

In recent years, coplanar waveguide (CPW) resonators have proven to be a unique tool for probing a wide variety of excitations in circuits and materials, including superconducting qubits [1], ferromagnetic spin ensembles [2], and magnons [3]. Due to their ultralow ohmic losses, they usually exhibit high quality factors, making them very sensitive to external perturbations. As their properties are, to a large extent, defined by their geometry and by the dielectric response of their environment, they make a promising candidate as probes for materials with exotic electronic properties. Transition metal oxide (TMO) heterointerfaces exhibit a variety of electronic and structural properties, including two-dimensional (2D) superconductivity [4], ferroelectric/magnetic orders [5], and negative capacitance [6]. Probably the most ubiquitous representative of transition metal oxides is SrTiO₃. Its chemical stability, lattice constant, and dielectric characteristics make it one of the standard substrates for the growth of high-quality crystalline thin films.

At low temperatures SrTiO₃ is characterized by an extremely high dielectric response ($\epsilon_r = 23\,000$ below 4 K [7–10]) originating from an incipient ferroelectric transition (quantum paraelectricity). The permittivity is affected by electric fields, and below 4 K it can be lowered by more than 1 order of magnitude for a field of about 2 MV/m [11,12]. The gate-tunability of the dielectric constant of SrTiO₃, particularly in thin-film form, has been used in applications of resonators incorporating SrTiO₃ for voltage-controlled microwave filters [13–18]. Beyond applications, one can also envision using superconducting resonators for sensing and exploring the dielectric properties of SrTiO₃ itself, similar to the application of superconducting cavities to probe qubits, spin ensembles, and magnetic excitations. The integration of CPW technology with oxide heterostructures is a novel approach towards studying these complex materials as a high-precision and device-oriented technique. However, it first

requires understanding of how the presence of the SrTiO₃ substrate affects the response of planar devices working at microwave frequencies.

Here, we use superconducting CPW resonators fabricated on single-crystal SrTiO₃ to explore the dielectric response of this quantum paraelectric material. As a reference, we fabricate a nominally identical resonator on single-crystal Al₂O₃ (sapphire). Since sapphire is a well-known standard substrate with low dielectric losses [19], we were able to perform a quantitative analysis on the dielectric response of the resonator fabricated on top of the SrTiO₃ sample under different excitations by direct comparison of the two resonators.

II. EXPERIMENTAL SETUP

A superconducting coplanar waveguide resonator is fabricated on top of a TiO₂-terminated single-crystal SrTiO₃(001) substrate ($5 \times 5 \times 0.5$ mm³). The CPW resonator is realized by a standard lithographic process followed by sputtering of a 135-nm-thick molybdenum-rhenium (Mo_{0.60}Re_{0.40}) film and lift-off in acetone. A cross-sectional schematic of the device is presented in Fig. 1(a). The CPW resonator is a 45- μ m-long line with a meander shape, and the central strip is 30 μ m wide, with a 10- μ m gap to the ground plane. The geometric parameters are chosen such that a significant impedance mismatch is created between the transmission line ($Z_0 = 50 \Omega$) and the resonator ($Z_{CPW} = 1 \Omega$ at 3.5 K, which acts as a mirror for microwave photons). This, together with the open end of the resonator, creates a $\lambda/2$ cavity. Figure 1(b) shows a top-view optical image of the device together with a schematic of the measurement setup. Our measurement scheme provides galvanic access to the central strip, so a dc bias can be easily applied with respect to the ground plane. Reflection measurements (S_{11}) are performed using a vector network analyzer, while the sample is always kept at a fixed temperature of 3.5 K.

We obtain information on the dielectric properties of the SrTiO₃ by direct comparison with an identical resonator, fabricated on top of a sapphire substrate (see Supplemental Material S1 [20]). As the inductance of both waveguides is the

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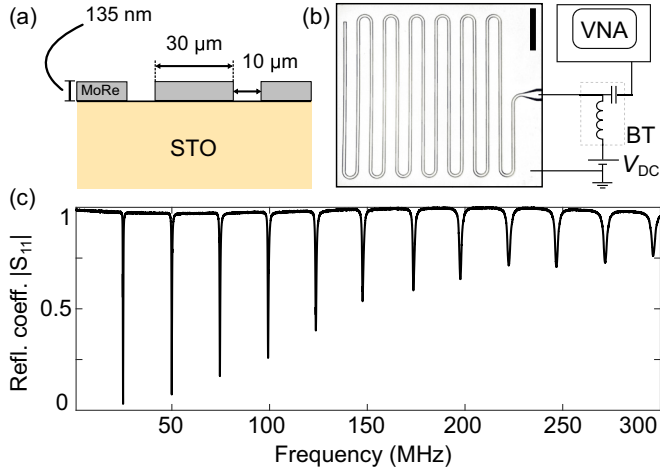


FIG. 1. Design of a superconducting CPW resonator as a probe of the dielectric response of SrTiO₃. (a) Cross-sectional schematic of the device. (b) Top-view optical image of the $\lambda/2$ CPW resonator with a schematic of the measurement setup. (VNA: vector network analyzer; BT: bias tee). (c) Reflection measurement $|S_{11}|$ taken at 3.5 K. Twelve resonance modes are visible in the 1–300-MHz range.

same, the ratio of the frequencies of the fundamental modes of the two resonators ($f_1^{\text{Al}_2\text{O}_3}$ and f_1^{STO}) allows us to extract the effective dielectric constant of the SrTiO₃ as follows:

$$\epsilon_{r,eff}^{\text{STO}} = \epsilon_{r,eff}^{\text{Al}_2\text{O}_3} (f_1^{\text{Al}_2\text{O}_3} / f_1^{\text{STO}})^2. \quad (1)$$

The effective dielectric constant accounts for the electric-field distribution through the vacuum above the substrate using a linear correction factor ($\epsilon_{r,eff} = \alpha \epsilon_r$). For dielectric constants in the range of the measured values for SrTiO₃ ($\epsilon_r = 2.000$ – 30.000), the correction factor α^{STO} is calculated to be $\alpha_{\epsilon_r=2000}^{\text{STO}} = 0.5000$ and $\alpha_{\epsilon_r=30000}^{\text{STO}} = 0.4998$ (using analytical expressions from [21]). Since sapphire has a much lower dielectric constant which is also anisotropic, $\alpha^{\text{Al}_2\text{O}_3}$ is calculated using a finite elements simulation and is found to be $\alpha^{\text{Al}_2\text{O}_3} = 0.6004$ (see Supplemental Material S2 [20]). The dielectric constant of SrTiO₃ is strongly anisotropic, where the different crystal orientations show a variation of ϵ_r higher than a factor of 2 [7]. This means that planar geometries will sense an effective dielectric constant arising from an average over the different crystal orientations, domain configurations, and regions with different intensity of electric field (see Supplemental Material [20] S2).

A reflection measurement $|S_{11}|$ of the cavity modes is presented in Fig. 1(c), where 12 resonance modes are observed in the range from 1 to 300 MHz. The fundamental resonance frequency is 25.04 MHz, corresponding to $\epsilon_r = 27\,400$, calculated from the observed frequency of the sapphire sample $f_1^{\text{Al}_2\text{O}_3} = 1.241$ GHz.

III. ELECTRIC-FIELD DEPENDENCE OF ϵ_r

The galvanic connection of the feedline to the CPW resonator enables us to apply a voltage difference between the central strip and the ground plane. The narrow 10- μm gap converts a relatively small voltage to an intense and

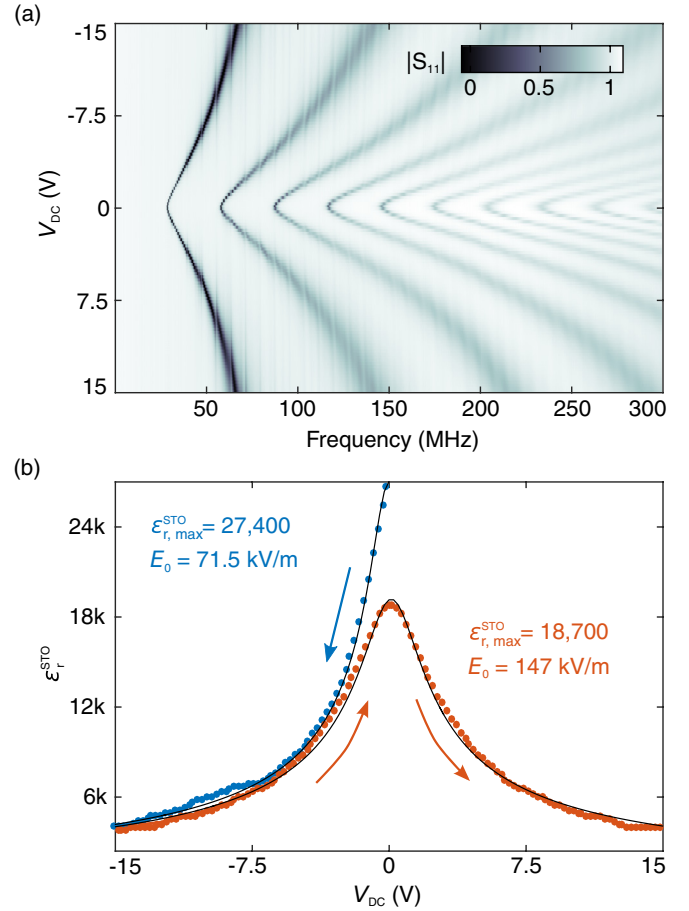


FIG. 2. dc voltage response of the resonator. (a) Color plot of $|S_{11}|$, showing the frequency shift of the cavity modes upon applying dc voltage in the ± 15 -V range. (b) Voltage dependence of ϵ_r of the SrTiO₃ substrate calculated from the frequency shift of the first mode of the CPW resonator. The blue data points are measured right after cooldown, while the red data points are taken after subsequent voltage sweeps. The black lines are fits to the data using Eq. (2).

localized average electric field (1 V \approx 100 kV/m) whose magnitude is further enhanced in the proximity of the CPW by the inhomogeneous density of electric-field lines (see Supplemental Material S2 [20]). A similar local gating scheme was used recently in transport experiments in oxide systems [22–24]. The modification of the local dielectric constant caused by the dc bias results in a change of the total capacitance of the resonator and a corresponding shift of its resonance frequencies. By tracking the resonance frequency as a function of applied voltage across the gap, we probe the voltage dependence of the dielectric properties of SrTiO₃ in a wide range of values.

In Fig. 2(a) we show a color plot of the reflection parameter $|S_{11}|$ as a function of dc voltage from -15 to 15 V. By applying -15 V to the central strip, the fundamental mode shifts from 25 MHz to 66 MHz, which corresponds to a tunability of 160%. By applying 30 V the fundamental frequency shifts to 90 MHz, resulting in a tunability of 260% (see Supplemental Material S3 [20]). The extracted dielectric constant as a function of the applied voltage is plotted in Fig. 2(b). We

calculated $\varepsilon_r^{\text{STO}}$ during the first voltage sweep just after the cooldown (black dashed line) and after subsequent sweeps in the ± 15 V range (orange solid line). There is an evident nonreversible modification of the dielectric constant at low bias after the application of the voltage, where $\varepsilon_r^{\text{STO}}(0)$ is reduced from the initial value of 27 400 to less than 19 000. This behavior, typically observed in SrTiO₃-based devices, is usually attributed to pinning of domain walls [25]. The electric-field dependence of the dielectric constant of SrTiO₃ at low temperatures can be described in first approximation by the Landau-Ginzburg-Devonshire theory [26,27]:

$$\varepsilon_r^{\text{STO}}(E) = 1 + \frac{\varepsilon_r^{\text{STO}}(0)}{[1 + (E/E_0)^2]^{1/3}}, \quad (2)$$

where E is the electric field, and E_0 is a parameter related to the tunability of the dielectric constant with electric field. During the first field sweep (black dashed line), we extracted a value for $E_0 = 71.5$ kV/m, whereas in subsequent sweeps (orange line), this value increased to $E_0 = 147$ kV/m, indicating a decrease in the polarizability of the SrTiO₃. In Sec. V, we provide a detailed analysis of this effect. We note that both these values are about 1 order of magnitude lower than what was reported in literature from measurements on bulk single-crystal SrTiO₃ with homogeneous electric fields [28]. This is due to the inhomogeneous distribution of the electric field, which is enhanced in the proximity of the central strip and the ground plane of the resonator. This allows us to affect the dielectric response at much lower voltages than those required in a double-capacitor geometry. It is worth mentioning, however, that despite the fact that we are probing the volume average of the properties of a strongly nonlinear material, the theoretical functional formulation still holds, supporting our analysis based on effective quantities.

IV. POWER AND FIELD DEPENDENCE OF MICROWAVE LOSSES

Due to their low intrinsic losses [29], superconducting MoRe resonators provide a good platform for characterization of the loss mechanisms of the surrounding dielectric environment. Dielectric losses are typically evaluated through the loss tangent of the substrate ($\tan \delta$), defined as the ratio between the imaginary (ε'') and real part (ε') of the relative permittivity,

$$\tan \delta \equiv \varepsilon''/\varepsilon', \quad (3)$$

which can be directly obtained from fitting the reflection coefficient from a single port cavity ($|S_{11}|$) with its definition:

$$|S_{11}| = \frac{2f_0/Q_{\text{int}} - \Delta f - 4i\pi(f - f_0)}{\Delta f - 4i\pi(f - f_0)}, \quad (4)$$

where Δf is the extracted FWHM of the resonance peak, Q_{int} is the internal quality factor, and f_0 its frequency. The loss tangent of the resonator ($1/Q_{\text{int}}$) usually incorporates both surface losses (originating from amorphous interfaces) and substrate losses. By comparison with the identical resonator fabricated on top of sapphire we can estimate the contribution of the radiation and quasiparticle losses to be less than 7×10^{-4} . We therefore use $1/Q_{\text{int}}$ and $\tan \delta$ interchangeably.

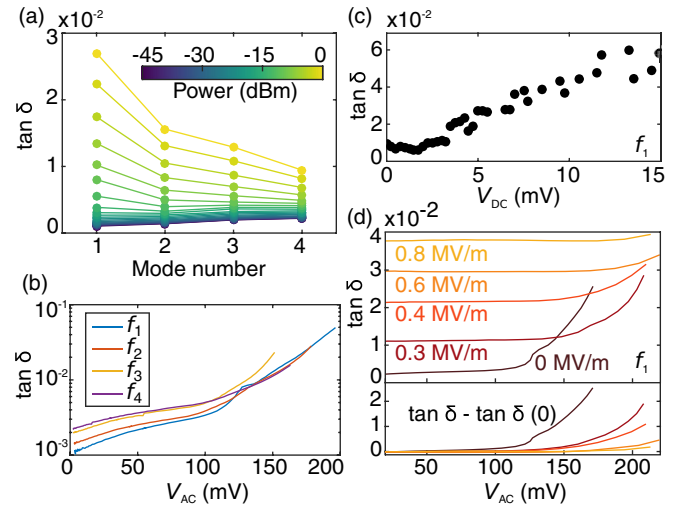


FIG. 3. Dielectric losses in SrTiO₃ probed by the CPW resonator: (a) $\tan \delta$ vs mode number at zero dc field measured at different rf powers (curves are 2 dBm apart). (b) $\tan \delta$ vs the ac voltage inside the cavity at resonance for the first four modes. (c) $\tan \delta$ vs dc voltage for the first mode of the resonator probed at fixed rf power. (d) Loss tangent for the first mode of the resonator as a function of the internal ac voltage for different values of the dc voltage. The bottom panel shows the overlapped curves, obtained by subtracting the value of $\tan \delta$ at minimum V_{AC} .

Figure 3(a) shows the loss tangent for the first four resonance modes for different input rf powers (P_{in}). Each curve corresponds to a different value of the rf power. For low values of P_{in} ($P_{\text{in}} < 10$ dBm), $\tan \delta$ for all modes is weakly affected by the input power and it increases with mode number. When increasing the input rf power, the losses become strongly affected by the ac field and their mode dependence is reversed, as lower modes seem to react more strongly to the input rf power.

In Fig. 3(b) we plot $\tan \delta$ of the first four modes, but now as a function of the internal ac voltage rather than input power. This is important because the different internal quality factors of the modes result in different amplitudes of the ac voltage inside the resonator for the same input power. At low ac voltages ($V_{\text{AC}} < 90$ mV), we observe a monotonic dependence of $\tan \delta$ on mode number: lower modes exhibit lower $\tan \delta$. This suggests that $\tan \delta$ is frequency dependent, similar to what was observed in [10] for gigahertz frequencies. In this range, all modes show a linear dependence on the ac voltage with similar slopes (see Supplemental Material S4 [20]). For higher amplitudes of the ac voltage ($V_{\text{AC}} > 90$ mV) we find an exponential increase of the losses for all four modes. In the high-power region, the peaks become progressively broader and we also see signatures of nonlinear damping. Details on the derivations and fits of the data are presented in Supplemental Material S4 [20].

The observed power dependence of $\tan \delta$ is not consistent with the typical behavior of dissipation in the two-level system (TLS) model, a common source of losses at the metal-substrate/metal-air interfaces, since such interfacial losses usually show an opposite saturation behavior [30,31]. This suggests that the measured $\tan \delta$ likely originates from substrate losses.

To better understand the loss mechanisms, in Fig. 3(c) we also investigate the dependence of $\tan \delta$ as a function of dc electric field extracted from the resonance measurements for the first dc sweep. The increase of $\tan \delta$ with V_{DC} is in agreement with the so-called quasi-Debye mechanism [32,33], which is the dominant dissipation channel in incipient ferroelectrics. This mechanism originates from the interplay between dc and ac components of the excitation. The dc field breaks the lattice symmetry, making the phonon spectrum field-dependent. The ac field acts as a time modulation of the phonon frequencies, which are continuously driven out of equilibrium with consequent energy dissipation. This increase has been already observed in SrTiO₃ single crystals [34], while it is in striking difference with several studies on thin films, where the application of an electric field leads to a decrease in $\tan \delta$ [14,35].

Figure 3(d) shows the ac-induced losses for different values of the dc voltage for the first resonance mode where, similarly to Figs. 3(a) and 3(b), two different regimes can be distinguished. It is possible to compare the losses related to the ac field at different dc bias by considering their rescaled values: $\tan \delta(V_{AC}) - \tan \delta(V_{AC} = 0)$, which are plotted in the bottom panel of Fig. 3(d). The presence of a dc field reduces the magnitude of the ac-induced losses, which suggests that the two dissipation channels are not fully independent.

In contrast to what is observed in thin films, we measured an increase of the losses with the magnitude of both ac and dc fields. According to Zubko and Vasil'ev [36], the ac and dc field dependence of the losses is determined to a large extent by the parameter ξ_S , which characterizes the quality of the crystal ($\xi_S \approx 0.02 - 0.06$ for high-quality single-crystal SrTiO₃ and $\xi_S > 0.5$ for thin films [10,37]). For values of $\xi_S < 0.25$, $\tan \delta$ should monotonically increase with both dc and ac electric fields, which is in agreement with our measurements.

V. DEPENDENCE OF ϵ_r AND $\tan \delta$ ON FIELD POLARITY

In Fig. 4, we explore the influence of voltage polarity on the hysteretic response of the dielectric properties of SrTiO₃. To do so, we followed the measurement sequences sketched in Fig. 4(a): starting from zero bias, we applied a voltage ramp up to a target value V_{max} and then back to $V = 0$, where the CPW resonator spectrum is acquired. This procedure is repeated for increasing values of V_{max} up to 15 V, always with the same polarity. We then perform a thermal cycle up to room temperature, while keeping the line grounded, to restore the initial sample conditions. We probed under three biasing schemes: positive, negative, and positive after negative (zero average).

Figure 4(b) shows that the value of ϵ_r at zero field is not symmetric with respect to the applied field polarity and, in particular, it is significantly affected only by negative biases. We thus link the origin of the reduction of permittivity to the presence of oxygen vacancies, since a negative bias can increase their concentration in the surroundings of the waveguide, where the dielectric environment is probed. This scenario is supported by the partial recovery observed when a positive voltage follows a negative one (yellow squares of

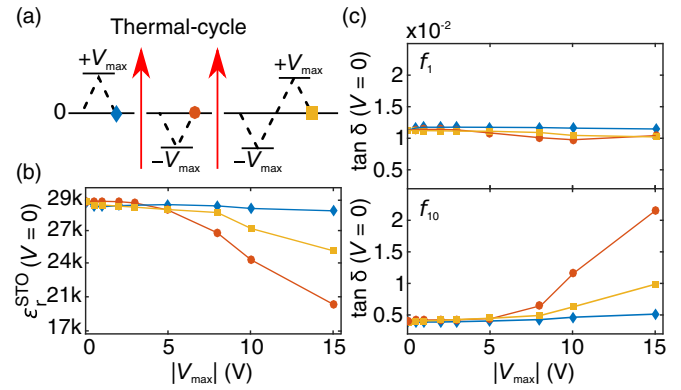


FIG. 4. Hysteretic effects in the dielectric response of SrTiO₃. (a) Diagram showing the biasing and measurement sequence used to characterize the dependence of ϵ_r^{STO} (b) and $\tan \delta$ (c) on the field polarity. Measurements are acquired at zero dc bias after the application of a given voltage difference (V_{max}) between the line and the ground plane. We kept a constant polarity for the different V_{max} values. After reaching $V_{max} = \pm 15$ V, a thermal cycle to 300 K restores the initial conditions of the sample. (Top: fundamental mode (f_1), bottom: mode f_{10} .)

Fig. 4). This explains the low tunability reported for CPW resonators realized on top of SrTiO₃ thin films, since even the very low defects concentration present at the surface of our substrate is already capable of lowering the dielectric response by more than 25% after the application of 15 V. Even though the cooldown conditions were kept unchanged, the values of ϵ_r and $\tan \delta$ at zero bias upon cooldown show a spread of about 2%. This value is negligible with respect to the observed variations and may be caused by different stable configurations of the oxygen vacancies at room temperature. This asymmetric response is also observed in the dielectric losses presented in Fig. 4(c). However, we note that the measured response depends unexpectedly on the mode number. As a general trend, a monotonic increase of the losses is observed, which is explained by a progressive accumulation of polarized defects for negative biases [33]. However, for low mode numbers and bias values below 10 V, the application of an electric field results in a decrease of $\tan \delta$. In the Supplemental Material S5 and S6 [20], we provide an extensive mapping of the losses showing the evolution of $\tan \delta$ as a function of the maximum applied voltage, mode number, and input power. In particular, upon having applied a dc voltage of -15 V and performing the same analysis as in Fig. 3(a), we observe a stronger dependence of $\tan \delta$ on mode number at low powers, whereas, for higher input powers, $\tan \delta$ of the first four modes shows a similar behavior.

We also extract the polarizability parameter E_0 and the horizontal shift of the maximum of ϵ_r^{STO} by fitting the ϵ_r^{STO} vs V_{DC} curves during the voltage sweeps (Supplemental Material S7 [20]). Finally, to support our interpretation of the fatigue mechanism, in Fig. S8 of the Supplemental Material we perform a measurement of the time evolution of ϵ_r^{STO} upon applying a small negative dc voltage [20]. The time response of the dielectric constant indicates the presence of slow relaxation dynamics, which is in accordance with the proposed model of drift of charged defects.

An advantage of the described method is that the superconducting cavity is fabricated directly on top of the SrTiO₃ and serves as a local probe of the underlying dielectric. In addition, the use of a reference sample facilitates the extraction of quantitative information on the dielectric properties of the material. This measurement technique is compatible with a wide variety of complex oxides, it enables local electrostatic gating, and it can serve as a platform for time-resolved measurements in the millisecond range.

VI. CONCLUSIONS

In conclusion, we realized a superconducting coplanar waveguide resonator directly on top of a single-crystal SrTiO₃(001) substrate and probed its dielectric properties as a function of dc voltage, rf power injected in the line, and mode number. The effective dielectric constant measured in the initial condition was $\sim 27\,000$, lowered below 4000 by applying a dc voltage of 15 V. The inhomogeneous distribution of the electric field in the resonator geometry allowed us to uncover an asymmetric response with respect to the field polarity of both ϵ_r and $\tan \delta$, with a progressive reduction of the resonator's tunability range, pointing towards the presence of charged defects, such as oxygen vacancies, as

the origin of the observed behavior. The narrow gap (10 μm) increases the sensitivity of the microwave response to effects on a length scale of the order of the gap size. By further shrinking its dimensions, a microwave cavity can act as a local probe capable of studying, for example, the dynamics and the dielectric behavior of single domain walls, characteristic of SrTiO₃ at low temperatures. Our results demonstrate the sensitivity of superconducting microwave cavities as probes of quantum matter and provide a robust background for future studies on oxide heterostructures probed by superconducting resonators.

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D.D. and N.M. contributed equally to this work.

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