

Ferroelectric switching and temperature-dependent electrical properties of $\text{K}_{0.7}\text{Na}_{0.3}\text{NbO}_3$ capacitor devices

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We performed the macroscopic electrical characterization of epitaxially grown, compressively strained $\text{K}_{0.7}\text{Na}_{0.3}\text{NbO}_3$ thin films (70 nm) on $\text{SrRuO}_3/(110)\text{TbScO}_3$ substrates, displaying monoclinic M_c domains. Hysteresis measurements revealed ferroelectric behavior up to at least 10^7 cycles at room temperature, with a polarization value ($2P$) of $25 \mu\text{C}/\text{cm}^2$. A double-peak switching behavior was observed, attributed primarily to domain-wall pinning/depinning and back switching, with a wake-up effect analogous to that reported in doped hafnium oxide and lead zirconate titanate systems. Temperature-dependent measurements (100 K–300 K) confirm ferroelectricity at low temperatures, while the double-peak behavior becomes less pronounced due to suppression of thermally activated back switching. Capacitance-voltage measurements reveal an increased dielectric response at higher temperatures, consistent with enhanced domain wall mobility. The asymmetry in coercive voltages additionally points to internal bias fields likely arising from electrode interfaces. Our results provide a comprehensive overview of the switching dynamics of strained $\text{K}_{0.7}\text{Na}_{0.3}\text{NbO}_3$ films and demonstrate ferroelectricity in the M_c phase.

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I. INTRODUCTION

As a lead-free alternative to high-performance piezoelectrics, $\text{K}_x\text{Na}_{1-x}\text{NbO}_3$ and derived materials in the form of ceramics [1–6], bulk single crystals [7,8], or thin films [9,10], have been extensively studied in regard to their crystal structure, phase transitions, and electrical properties across a wide range of compositions. Just as in its lead-based perovskite counterparts, $\text{K}_x\text{Na}_{1-x}\text{NbO}_3$ exhibits a rich phase diagram [11] characterized by multiple ferroelectric transitions and diverse degrees of freedom of its spontaneous polarization, which is of great interest for technological applications [7].

After Noheda *et al.* (1999) [12] first reported the presence of a monoclinic phase in $\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$, subsequent experimental and theoretical studies linked this phase to enhanced piezoelectricity via polarization rotation within a mirror plane [13–15], which led to growing interest in piezo- and ferroelectric materials with monoclinic structures. The rotation mechanism in monoclinic phases allows variable out-of-plane polarization components, offering multiple accessible states that are particularly interesting for use in ferroelectric

switching applications such as transistors, memristors, or energy storage [16–19].

Lattice-mismatch strain, mediated by heteroepitaxial growth of thin $\text{K}_x\text{Na}_{1-x}\text{NbO}_3$ films, for instance on rare-earth scandate substrates (REScO_3), enables tuning of the phase transition temperatures and stabilizes the monoclinic phase at room temperature, as evidenced by x-ray diffraction and piezoresponse force microscopy (PFM) [20–24]. Under compressive strain, epitaxial films exhibit periodic patterns of M_c domains at room temperature [25,26], which can be switched locally by applying a bias with a PFM tip [27].

However, the growth of high-quality ferroelectric epitaxial $\text{K}_x\text{Na}_{1-x}\text{NbO}_3$ films and, in particular, the fabrication of reliable capacitor structures remains technically challenging. Several works have reported ferroelectric switching in $\text{K}_x\text{Na}_{1-x}\text{NbO}_3$ -based thin films prepared by pulsed laser deposition (PLD), sol gel, and RF sputtering methods, but their integration into capacitor devices is hindered by issues such as low film quality, leakage, high coercive fields, and large film thickness [28–34]. Due to the volatility of the alkali metals and oxygen, only the growth by metal-organic vapor phase epitaxy (MOVPE) has provided sufficient control of the chemical composition of the $\text{K}_x\text{Na}_{1-x}\text{NbO}_3$ films. These MOVPE-grown films show well-ordered monoclinic M_c stripe domain patterns, yet their integration into vertical capacitor devices has not been widely explored. Few studies of the electrical performance of devices based on strained thin $\text{K}_x\text{Na}_{1-x}\text{NbO}_3$ films with a monoclinic phase have been performed so far and, thus, key ferroelectric properties, such as polarization, capacitance, fatigue behavior, and temperature dependence, remain largely unknown. This lack of reliable

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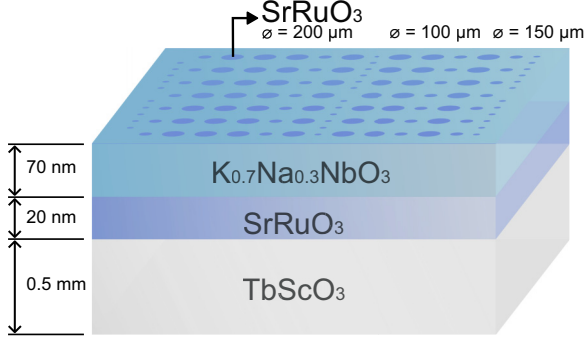


FIG. 1. Scheme of the 70 nm $\text{K}_{0.7}\text{Na}_{0.3}\text{NbO}_3/\text{SrRuO}_3/$ (110) TbScO_3 device with SrRuO_3 top electrodes.

device fabrication limits the technological exploitation of strained $\text{K}_x\text{Na}_{1-x}\text{NbO}_3$ thin films.

In this work, we successfully demonstrate the ferroelectric switching in macroscopic $\text{K}_x\text{Na}_{1-x}\text{NbO}_3$ -based capacitor devices. In particular, we present a detailed electrical characterization of compressively strained, (001) $_{pc}$ -oriented $\text{K}_{0.7}\text{Na}_{0.3}\text{NbO}_3/\text{SrRuO}_3/(110)\text{TbScO}_3$ thin films displaying a stable M_c phase. Our data indicate that the devices exhibit stable ferroelectricity up to 10^7 cycles without showing any sign of fatigue. We furthermore discuss the possible mechanisms of the observed temperature-dependent switching/back-switching behavior of the domains.

II. EXPERIMENT

The overall epitaxial layer stack is depicted in Fig. 1, and details about the crystallographic orientation can be found in von Helden *et al.* (2019) [26]. An initial 20 nm thick, strained SrRuO_3 layer, acting as bottom electrode, was deposited on a (110)-oriented TbScO_3 substrate by PLD. The (110) substrate orientation corresponds to the (100) $_{pc}$ plane of the pseudocubic, primitive perovskite unit cell. The growth was conducted at a substrate temperature of 750 °C, an oxygen partial pressure of 0.15 mbar, a KrF laser (248 nm) fluence and frequency of 50 mJ and 5 Hz, respectively, and a target-to-substrate distance of 55 mm. A slight off orientation of the TbScO_3 growth surface of 0.1° leads to a well-ordered step-and-terrace structure after annealing in pure oxygen.

The 70 nm $\text{K}_{0.7}\text{Na}_{0.3}\text{NbO}_3$ film was then epitaxially grown by liquid-delivery spin MOVPE at 730 °C. $\text{K}(\text{tmhd})$, $\text{Na}(\text{tmhd})$ [tmhd = 2, 2, 6, 6-tetramethyl-3,5-heptanedione], and $\text{Nb}(\text{EtO})_5$ [(EtO) $_5$ = pentaethoxide] dissolved in dry toluene, were used as metal-organic source materials of K, Na, and Nb, respectively. More details are found in Schwarzkopf *et al.* (2012) [35]. Subsequently, thin SrRuO_3 top electrodes with an estimated thickness of 20 nm and diameters ranging from 100 to 200 μm were deposited on the film via PLD using a shadow mask at 690 °C substrate temperature, oxygen partial pressure of 0.15 mbar, a laser fluence and frequency of 50 mJ and 5 Hz, respectively, and target-to-substrate distance of 55 mm. The substrate temperature is reduced in comparison to the one used for the bottom electrode deposition in order to minimize loss of the volatile alkali metals by diffusion into the SrRuO_3 layer or outgassing. The whole stack forms

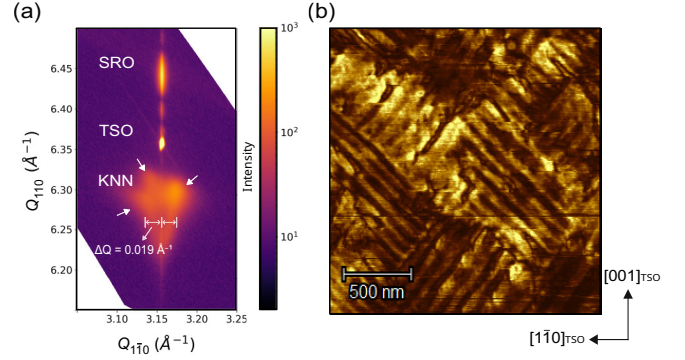


FIG. 2. (a) Reciprocal space map around the asymmetric 620 TbScO_3 reflection, where intensity is given in counts/s. Out-of-plane alignment between the TbScO_3 substrate (intersect at $Q_{110} = 6.35 \text{ \AA}^{-1}$), the SrRuO_3 bottom electrode (intersect at $Q_{110} = 6.44 \text{ \AA}^{-1}$) and the $\text{K}_{0.7}\text{Na}_{0.3}\text{NbO}_3$ film peaks is observed. The domain walls lead to additional peaks marked by the arrows, and the domain periodicity of $0.019 \text{ \AA}^{-1}$ is marked by the white bars. (b) $2 \times 2 \mu\text{m}^2$ lateral piezoresponse force microscopy amplitude image.

an epitaxial $\text{SrRuO}_3/\text{K}_{0.7}\text{Na}_{0.3}\text{NbO}_3/\text{SrRuO}_3$ capacitor fully strained on the TbScO_3 substrate [Fig. 1(a)]. We note that attempts to fabricate capacitor structures using alternative metal electrodes (Pt and Au) resulted in high leakage currents and poor ferroelectric response. In contrast, epitaxial SrRuO_3 enabled reliable capacitor performance.

Structural characterization was performed using a high-resolution x-ray diffractometer (9 kW SmartLab, Rigaku) with $\text{Cu K}\alpha_1$ radiation ($\lambda = 1.54056 \text{ \AA}$). Ferroelectric domain patterns of different scanned regions were visualized by the lateral PFM mode of an Asylum MFP3D-instrument with dual AC resonance tracking (DART).

A ferroelectric tester (TF Analyzer 3000, aixACCT Systems) was employed to obtain the polarization hysteresis P - V loops (dynamic hysteresis measurements, DHM) and fatigue measurements. These were performed by applying triangular waveform pulses, with a frequency of (i) 7 kHz for Figs. 3 and 7, and (ii) 1.5 kHz for Fig. 4, all with a delay time of 300 ns between consecutive pulses. Temperature dependent positive up, negative down (PUND), and capacitance-voltage (C - V) measurements were conducted on a Lake Shore Cryogenic CPX-VF probe station with liquid nitrogen as a coolant, which is described in detail by Lancaster *et al.* (2022) [36]. Voltage pulses were applied using a Keithley 4200A-SCS parameter analyzer with 4225 pulse measurement units, where triangular pulses were applied at 7 kHz and 5 μs delay time between pulses.

III. RESULTS

A. Structural characterization

PFM images [Fig. 2(b)] reveal the well-established locally periodic ferroelectric domain patterns of the M_c phase. The domains arrange themselves into four variants of superdomains, each with a unique net in-plane polarization vector. A detailed structural analysis of this domain arrangement is provided in von Helden *et al.* (2018) [25].

The reciprocal space map [Fig. 2(a)] confirms pseudomorphic epitaxial growth of both the $\text{K}_{0.7}\text{Na}_{0.3}\text{NbO}_3$ and SrRuO_3 layers on the substrate, as evidenced by the vertical alignment of their peaks, labeled KNN, SRO, and TSO, respectively. A $\theta - 2\theta$ scan is provided in the Supplemental Material (Fig. S1) [37]. In samples thinner than 60 nm, pronounced satellite peaks arising from the periodicity of M_c stripe domains within each of the superdomains appear [25]. Investigations of the effect of film thickness show an onset of elastic relaxation at thicknesses above 50 nm [38]. In our case, the 70 nm film is fully strained, although the relatively high thickness leads to smaller superdomains and larger period length of the contained M_c domains. The low number of periods per superdomains effectively results in a lower long-range order and, thus, to less pronounced satellite peaks, as also discussed in Wang *et al.* (2022) [38]. As seen in the reciprocal space map, only the first order satellite peaks are visible [highlighted by the white arrows in Fig. 2(a)]. The monoclinic splitting of the peaks is such that the rightward satellite peaks (at higher $Q_{1\bar{1}0}$) overlap. The two leftward satellite peaks are well separated and probe two of four superdomains that have parallel in-plane net polarization as seen in the lateral PFM image [Fig. 2(b)]. The domain periodicity extracted from PFM images corresponds to a reciprocal space periodicity of $0.019 \text{ \AA}^{-1}$, in agreement with the spacing between the main film peak and the additional diffuse satellite peaks, marked in Fig. 2(a).

B. Room-temperature switching and cycling-dependent evolution

At room temperature, polarization-voltage (P - V) and current-voltage (I - V) curves measured at 7 kHz [Fig. 3(a)] show the ferroelectric behavior of the devices, as evidenced by the peaks in the current and the resulting hysteresis of the P - V loop. The I - V curves feature two distinct switching peaks on each voltage side, which we refer to as P1-P4, from positive to negative bias. For the initial cycle, on the positive side, P1 and P2 occur at voltages of 1.11 V (159 kV/cm) and 0.37 V (53 kV/cm), respectively, while on the negative side, P3 and P4 appear at -0.52 V (-74 kV/cm) and -1.47 V (-210 kV/cm). Electrical field values (in kV/cm) are nominal averages calculated assuming a homogeneous vertical field distribution, i.e., no space or leakage charges in the layer. P1-P2 and P3-P4 form a characteristic “double-peak” signature, respectively.

The extracted polarization values at zero voltage are $P^+ = +10.68 \mu\text{C}/\text{cm}^2$ and $P^- = -14.28 \mu\text{C}/\text{cm}^2$, which are in the same range as for previously reported values for much thicker (200 nm–30 μm) $\text{K}_{0.7}\text{Na}_{0.3}\text{NbO}_3$ films [29,31–33,39,40]. Here and throughout, we define $2P \equiv |P^+| + |P^-|$. We note, however, that asymmetric leakage currents, particularly for negative polarity, contribute to the apparent polarization in conventional P - V loops. Additionally, domain back switching leads to different values for polarization at zero voltage when extracted from dynamical hysteresis measurements and remanent polarization extracted from PUND measurements. Because we are investigating the back-switching behavior of our devices, their contribution is not

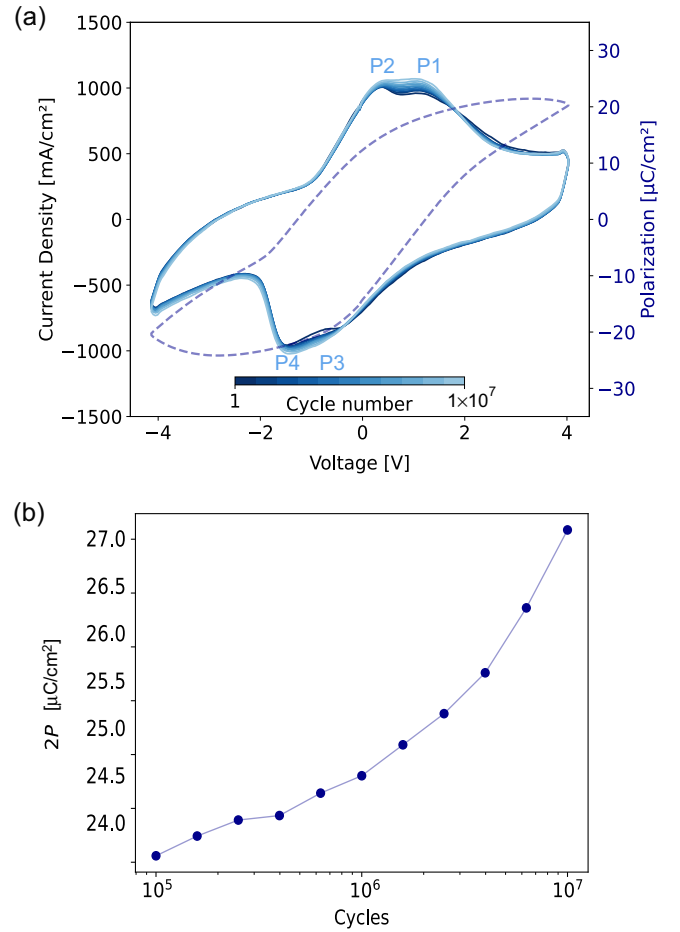


FIG. 3. Room temperature electrical measurements for the 70 nm $\text{K}_{0.7}\text{Na}_{0.3}\text{NbO}_3/\text{SrRuO}_3/(110)\text{TbScO}_3$ film measured at 7 kHz. (a) Polarization-voltage (P - V , dashed, dark blue) hysteresis loop for the initial cycle, and current-voltage (I - V , blue shades) dependence on cycling, showing characteristic ferroelectric switching with labeled peaks P1–P4. (b) $2P$ extracted from hysteresis measurements show the endurance of the polarization over 10^7 switching cycles, with a gradual increase in polarization. No fatigue was observed in the devices.

excluded. Nevertheless, for comparison, the room temperature PUND-derived P - V loop is shown in the Supplemental Material (Fig. S2) [37]. The extracted remanent polarization value ($2P_r$) is of $15.60 \mu\text{C}/\text{cm}^2$, considerably lower than that of $2P$ for dynamical hysteresis measurements.

An increase in the polarization [Fig. 3(b)] and a reduction of peak splitting [Fig. 3(a)] can be observed with bipolar ferroelectric cycling of the devices. The devices show no indication of degradation up to 10^7 cycles and a negligible increase in leakage currents. Additionally, hysteresis measurements of pristine devices with opposite polarity, starting with either a positive pulse in Fig. 4(a) or a negative pulse in Fig. 4(b), were performed at a lower frequency (1.5 kHz), since the back-switching peaks are more pronounced. These measurements show that the peaks closer to $V = 0$ V only appear once the device has been prepoled in the corresponding direction.

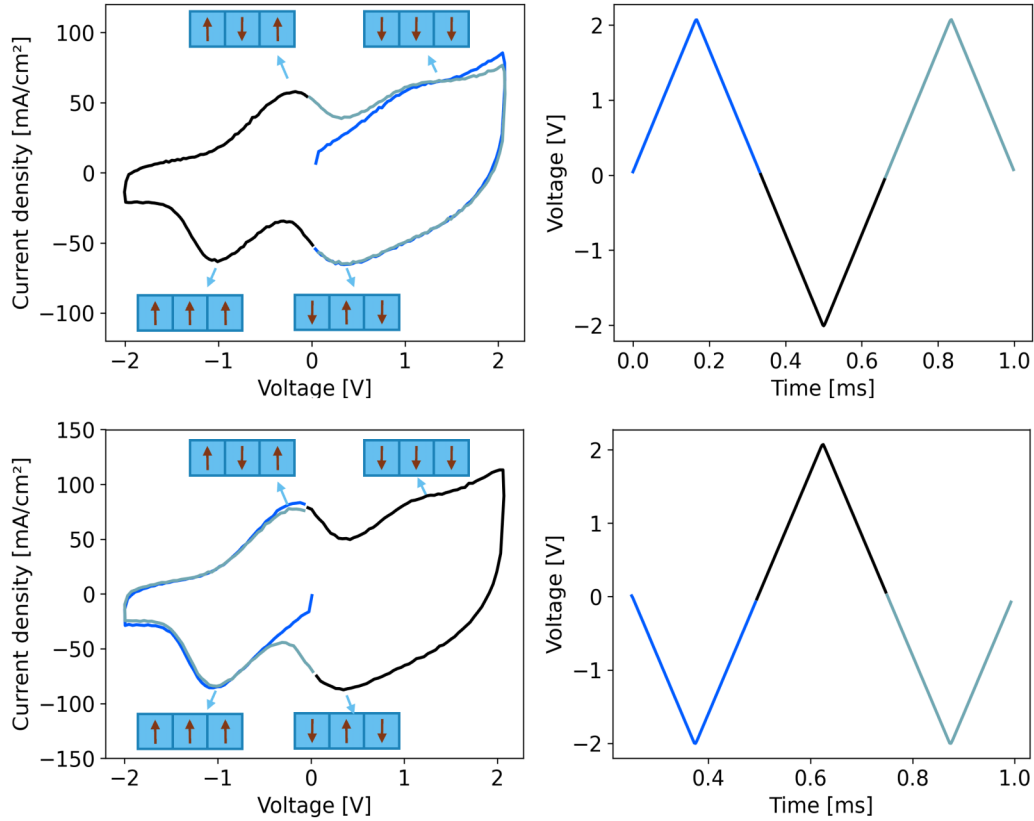


FIG. 4. Switching measurements of $\text{K}_{0.7}\text{Na}_{0.3}\text{NbO}_3/\text{SrRuO}_3/(110)\text{TbScO}_3$ devices at 300 K and frequency of 1.5 kHz. The measurements reveal double-peak behavior present only once the device has been prepoled, consistent with domain-wall pinning/depinning rather than transient charge-trapping effects. The insets provide simplified sketches of domain configurations at the switching peaks, illustrating the evolution of domain states during the reversal process.

C. Temperature-dependent switching

We investigated the switching behavior of our devices from 100 K to 300 K (Fig. 5). PUND-derived I - V curves are shown in Fig. 5(a). The curve extraction and differences between the curves measured by PUND and those measured by dynamical hysteresis measurements are described in the Supplemental Material [37]. In Fig. 5(a), it can be seen that the current densities of P2 and P3 reduce with decreasing temperature, in contrast to those for P1 and P4, which are almost independent of temperature. All peaks show a drift of their peak position with varying temperature. The ratio of peak intensities at the peak maximum (P1/P2 and P4/P3, i.e., between the dominant peak and the peak at lower voltage) decreases as temperature increases [Fig. 5(b)]. As expected, leakage current also decreases at lower temperatures.

Corresponding P - V loops are presented in the Supplemental Material (Fig. S3) [37]. At 100 K, the loop is noticeably tilted and, as the temperature increases, the loops become less tilted, so the saturation polarization (P_{sat}) increases. The tilting is understood as a wider distribution of the coercive field at lower temperatures, i.e., broader switching current peaks [41]. Additionally, the ratio between the two current peaks on each voltage side in the corresponding I - V curves [Fig. 5(a)] determines the fraction of domains that remain switched at $V = 0$ V.

Figure 6 presents the C - V curves measured at different temperatures, displaying the characteristic counterclockwise,

hysteretic, butterfly shape for ferroelectric materials. Each loop shows two capacitance maxima at the coercive voltages, where dielectric susceptibility (and also permittivity) is highest, just before domain reversal. Coercive voltage values extracted from the C - V curve at room temperature are $-0.81/0.62$ V, different from the values extracted from DHM or PUND measurements. This discrepancy is common in ferroelectric materials, particularly when the current peaks are not sharp, but rather distributed for two different peaks and across a voltage range [42].

D. Frequency-dependent switching

To further probe the switching dynamics, we measured I - V curves across a range of frequencies, from 1 to 20 kHz (Fig. 7). At low frequencies, the switching current peaks (P1–P4) appear sharp and well defined, while at higher frequencies they broaden and shift toward larger absolute voltages. Figure 7 also displays the extracted peak positions as a function of frequency. All peaks show a shift away from the origin, resulting in increased coercive voltage with frequency. Additionally, the switching current amplitude increases for P1 and P4 and decreases for P2 and P3. Therefore, the larger coercive voltages observed in Fig. 3(a), measured at 7 kHz, compared with those in Fig. 4, measured at 1.5 kHz, are a consequence of this frequency dependence.

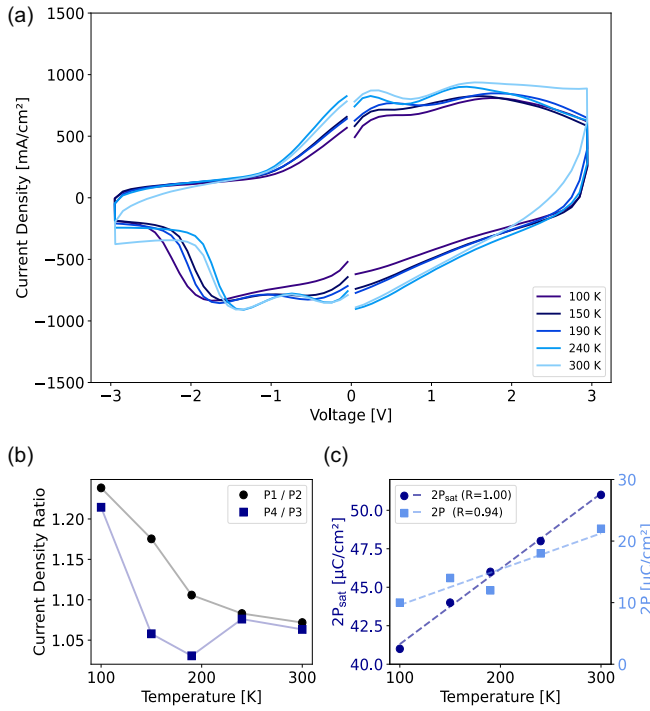


FIG. 5. (a) Temperature-dependent I - V curves highlighting the evolution of switching behavior across temperatures from 100 K to 300 K. (b) Ratio of switching peak current densities (P1/P2 and P4/P3) as a function of temperature, indicating asymmetry reduction at lower temperatures. (c) Development of P_{sat} (dark blue) and $2P$ (light blue) with temperature. While only a small temperature dependence of $2P$ can be observed, P_{sat} shows a linear increase with temperature.

IV. DISCUSSION

The origin of the double-peak behavior observed in our hysteresis measurements requires careful interpretation. Similar peak splitting has been observed in other ferroelectric thin films, such as $\text{PbZr}_x\text{Ti}_{1-x}\text{O}_3$ and HfO_2 -based ferroelectrics

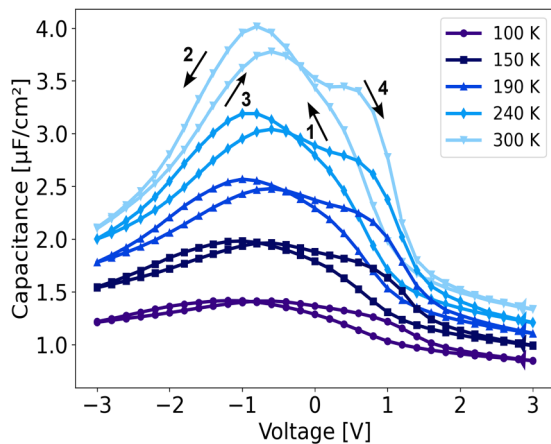


FIG. 6. Capacitance-voltage (C - V) characteristics for various temperatures, with labeled peaks (P1-P4) corresponding to the switching processes. Butterfly peaks increase with temperature, suggesting enhanced domain switching.

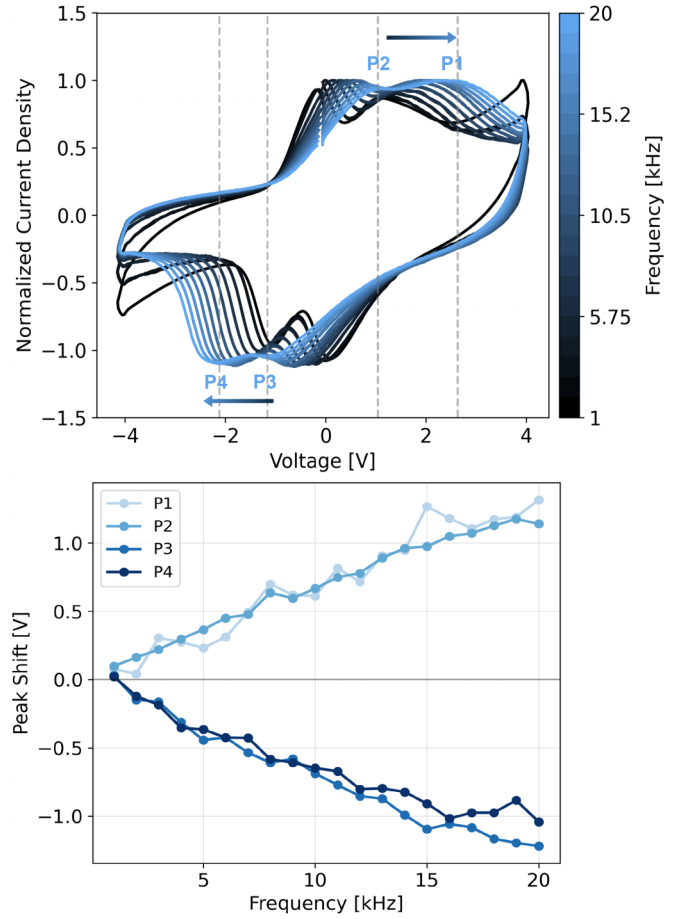


FIG. 7. Frequency dependence of I - V characteristics of $\text{K}_{0.7}\text{Na}_{0.3}\text{NbO}_3/\text{SrRuO}_3/(110)\text{TbScO}_3$ 70 nm thin films with SrRuO_3 top electrodes.

[43–46], and has been related to (i) trapped charged defects (e.g., oxygen vacancies) that accumulate at interfaces and create an internal bias field, (ii) domain wall pinning/depinning causing domain reorientation, or (iii) field-induced phase transitions from a nonferroelectric to a ferroelectric phase. Our results allow us to evaluate each possibility systematically.

In principle, a field-induced phase transition from a nonferroelectric to a ferroelectric phase could account for the double-current peaks. However, in our epitaxial films, neither PFM nor the reciprocal space maps indicate the presence of a nonferroelectric phase, which would be evidenced by a different domain pattern in the PFM images or by additional diffraction peaks in the reciprocal space map. The absence of such features points to extrinsic effects as the cause for the double-peak behavior.

Localized charge carriers can be trapped at defects or interfaces during a voltage pulse and subsequently detrapped under the opposite bias. This mechanism induces an internal bias that superimposes on the external field, which could also lead to peak splitting. Charge trapping results in a shift in peak position with frequency, as carriers have more or less time to be released. Additionally, the emission rate of trapped charges also reduces at low temperatures due to slower dynamics. Both tendencies are qualitatively consistent with the slower sweep and cooling experiments presented here. A contribution

from charge trapping can therefore not be entirely excluded. However, several key observations point toward domain-wall dynamics as the dominant mechanism, as will be explained below.

The switching-cycle measurements in Fig. 4 suggest that the double-peak behavior is primarily due to domain-wall pinning/depinning rather than charge trapping. The intermediate peaks P2 and P3 appear only once the device has been previously poled in the corresponding switching polarization direction, which is characteristic of domain back switching, where domains aligned during the initial pulse can partially revert as the external field is reduced, leading to the additional peaks. In the first pulse, only forward switching, but no initial back-switching peak, is observed. By reducing the external electric field back to zero, a fraction of the domains switch back before the coercive voltage of opposite sign is reached. The second pulse produces a similar partial back switching. Such systematic back switching is related to domain-wall dynamics, where pinned domains become unstable once the external field is lowered, in agreement with previous reports for HfO₂-based ferroelectrics [41].

With bipolar cycling [Fig. 3(a)], the double-peak behavior is gradually suppressed. This kind of wake-up effect is consistent with the redistribution of trapped charges and reduction of internal bias fields under repeated polarization reversals. This effect lowers the barriers that initially pin some of the domains. As a result, domain-wall mobility improves, the two peaks (P1/P2 and P3/P4) merge into one, and the polarization at zero voltage increases, reflecting a more uniform switching once an increasing amount of domains are depinned under the applied electric field. The absence of significant leakage current increase with cycling also rules out the possibility of a trapping-dominated mechanism as the main cause for the double-peak behavior.

Temperature-dependent measurements further reinforce this model. Cooling the devices from 300 K down to 100 K (Fig. 5) reduces thermal activation and hinders domain-wall depinning. As a result, domains that have switched during the initial pulse are more likely to remain pinned and do not revert when the external field is removed. Consequently, the low-voltage back-switching peaks (P2 and P3) become less pronounced at low temperatures. The higher-voltage peaks (P1 and P4), associated with the most easily switchable domains, remain visible. This indicates that a larger portion of domains remains aligned when the external field is reduced to zero. This behavior strongly indicates that back switching is thermally activated, in agreement with the model proposed by Mehmood *et al.* (2020) [41].

The temperature dependence of $2P_{\text{sat}}$ and $2P$ can be similarly understood due to domain-wall-mediated switching. At low temperatures, the higher activation barriers of pinned domain walls slow both depinning and the subsequent domain-wall motion, so that not all domains can fully switch within the applied pulse width. This leads to a reduction of both $2P_{\text{sat}}$ and $2P$. At the same time, reduced thermal activation suppresses back switching at zero field, as evidenced by the changing ratio between the forward-switching peaks (P1 and P4) and the back-switching peaks (P2 and P3). The combined effects of incomplete switching and hindered back switching result in the trends observed in Fig. 5.

In addition to ferroelectric switching, ferroelastic switching is also expected to contribute to the electrical response of our films. In the M_c phase, the film displays four ferroelastic domain variants, and high electric fields can modify their relative stability, leading to their reorientation or redistribution within the superdomains. Recent phase-field simulations of (K,Na)NbO₃ films confirmed that the polarization switching pathway is strain coupled, reporting that in-plane electric fields can drive in-plane polarization switching and ferroelastic 90° domain-wall motion. Under a top electrode, and assuming complete screening of surface charges, the switching process takes place by nucleation on these 90° domain walls at the thin film surface and their subsequent propagation [47]. Nevertheless, even though strain plays a significantly role in the switching mechanism, the evolution of the I - V curves with cycling [Fig. 3(a)] and the merging of peaks P2 and P3 indicate that the mechanism governing the back switching in our devices is electrostatic in nature.

A suppression of back switching is also observed when increasing the measurement frequency, as evidenced by Fig. 7. As the cycle period becomes shorter, domains have less time to revert, leading to a slight decrease in the amplitude of P2 and P3, while the forward-switching peaks P1 and P4 grow in intensity. In addition, all four peaks shift toward higher voltages with frequency, reflecting the increase of coercive voltage expected from nucleation-limited switching kinetics [48].

Therefore, while cycling enhances mobility by removing pinning barriers, cooling restricts mobility by reducing the thermal energy required for depinning. Increasing the measurement frequency, on the other hand, does not change the intrinsic domain-wall mobility but limits the available time for reversal, so that back-switching peaks (P2 and P3) are suppressed while the forward-switching peaks (P1 and P4) dominate. By analyzing these effects, we demonstrate that ferroelectric switching (and back-switching) behavior in strained M_c -phase K_{0.7}Na_{0.3}NbO₃ films is governed by domain-wall pinning/depinning processes, and strongly dependent on the thermal activation necessary to move domain walls, cycling history, and measurement timescale. For complete verification, other techniques such as *in situ* TEM would be helpful to support our model, such as in Grimley *et al.* (2016) [49].

The temperature effect on domain wall mobility is also observed in our C - V measurements, where at low temperatures the butterfly loops flatten and broaden, consistent with increased effective coercive electric field and restricted domain wall motion (see Fig. 6). Regarding interface effects, the asymmetry in peak height and position with respect to the voltage axis suggests that an internal bias field is present, likely from distinct thin film/electrode interfaces or defects. The former could be the case for our samples, since, even though bottom and top electrode are the same material, they have different interfaces due to depositions using different growth conditions (500 °C vs 750 °C, with differing O₂ partial pressures; see the Experiment section), leading to variations in composition, vacancy concentration, and interface termination. This built-in field also leads to one polarity switching more readily than the other, resulting in different coercive voltages for positive or negative bias applied, an effect also

observed in PFM studies [27]. More symmetric switching would likely require identical electrode interfaces.

Finally, the limited success in previous attempts to produce reliable $K_xNa_{1-x}NbO_3$ capacitors using other electrode materials, such as Au and Pt, highlights the importance of interface engineering. The requirement of epitaxial, isostructural electrodes such as $SrRuO_3$ points to interface-related leakage as a central challenge, potentially involving not only structural mismatch but also band alignment and defect-mediated conduction. These issues need systematic investigation, ideally combining electrical testing with *in situ* structural probes such as TEM [49], to disentangle the contribution of band bending, vacancy accumulation, and domain-wall pinning.

V. CONCLUSION

We have demonstrated stable ferroelectric switching in macroscopic capacitors based on 70 nm thick epitaxial $K_{0.7}Na_{0.3}NbO_3$ films exhibiting the monoclinic M_c phase. Using epitaxial $SrRuO_3$ top and bottom electrodes, we were able to overcome previous challenges of high leakage in such films. At room temperature, these devices exhibit polarization at zero voltage of around $20 \mu C/cm^2$, comparable to much thicker $K_xNa_{1-x}NbO_3$ -based films. No degradation of the ferroelectric behavior or increase of leakage is present over 10^7 cycles of polarization inversion. A characteristic double-peak behavior is observed for pristine devices and evolves into a single peak with cycling, representing a wake-up effect. This double-peak response is attributed primarily to

domain-wall pinning/depinning rather than charge-trapping or field-induced phase transitions, as supported by the suppression of back-switching peaks at low temperatures and their appearance only once the device has been prepoled.

The combination of film thickness, switching voltages below 2 V, and cycling endurance above 10^7 cycles make such devices promising as candidates for next-generation, lead-free nonvolatile memories, even though both (i) further work to confirm the suggested model, and (ii) stability of the polarization state, i.e., reduction of back switching, are necessary.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available because they are owned by a third party and the terms of use prevent public distribution. The data are available from the authors upon reasonable request.

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