

Low leakage current in heteroepitaxial $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ ferroelectric films on GaN

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Wurtzite (Al,Sc)N ferroelectrics are attractive for microelectronics applications due to their chemical and structural compatibility with wurtzite semiconductors, such as GaN and (Al,Ga)N. However, the leakage current in epitaxial stacks reported to date should be reduced for reliable device operation. Here, we demonstrate low leakage current in epitaxial $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films on GaN with well-saturated ferroelectric hysteresis loops that are orders of magnitude lower (i.e., 0.07 A cm^{-2}) than previously reported films ($1\text{--}19 \text{ A cm}^{-2}$) having similar or better structural characteristics. We also show that, for these high-quality epitaxial (Al,Sc)N films, structural quality (edge and screw dislocations), as measured by diffraction techniques, is not the dominant contributor to leakage. Instead, the small leakage in our films is limited by thermionic emission across the interfaces, which is distinct from the large leakage due to trap-mediated bulk transport in the previously reported (Al,Sc)N films. To support this conclusion, we show that $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ on lattice-matched $\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ buffers with improved structural characteristics but higher interface roughness exhibit increased leakage characteristics. This demonstration of low leakage current in heteroepitaxial (Al,Sc)N films and understanding of the importance of interface barrier and surface roughness can guide further efforts toward improving the reliability of wurtzite ferroelectric devices.

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I. INTRODUCTION

Switchable spontaneous polarization in ferroelectric materials has enabled unique device capabilities in microelectronics, such as nonvolatile ferroelectric random-access memory (FeRAM) and field effect transistors (FeFET) and microelectromechanical (MEMS) systems [1–3]. Desired material properties and characteristics vary depending on applications, but the process compatibility to existing semiconductor fabrication lines, large and robust remanent polarization, and low leakage current are preferable to realize reliable and cost-competitive devices. Recent efforts have developed emerging ferroelectrics,

such as fluorite HfO_2 -based materials [4,5] and wurtzite AlN- and ZnO-based materials [6–12]. These ongoing studies on ferroelectric materials have the potential to enable next-generation microelectronics.

Among these ferroelectric materials, wurtzite nitrides are promising candidates for such devices due to their attractive piezoelectric and ferroelectric properties and process compatibility with both commercially relevant III-N semiconductors and Si. Wurtzite (Al,Sc)N shows an improved piezoelectric response compared with pristine AlN [13], robust ferroelectricity with large spontaneous polarization [6,8], and submicrosecond polarization switching [14,15]. Indeed, various devices using wurtzite ferroelectrics, such as ferroelectric tunnel junctions [16], ferroelectric high electron mobility transistors (HEMTs) [17], and two-dimensional FETs [18], have been demonstrated. Wurtzite nitrides are suitable for these devices due to the low deposition process temperature ($<400^\circ\text{C}$) compatible with Si microelectronics, and the lack of relatively mobile oxygen ions that cause fatigue and associated reliability concerns in many oxide ferroelectrics [19,20].

The leakage current of wurtzite nitride ferroelectrics is a critical issue for memory applications, allowing undesired

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charge injection through the ferroelectric gate [17]. Many studies on this material class show nonsaturated polarization–electric field hysteresis loops due to leakage current contributions, especially for epitaxial (Al, Sc)N/GaN stacks [21–25] that are essential for a ferroelectric HEMT [17]. It is surprising that the leakage currents reported for such epitaxial stacks are far greater than those reported for nonepitaxial stacks of similar compositions [6,15,21–23,25,26]. The origins of leakage current in (Al, Sc)N are not fully understood yet, but dislocation density [24], impurities associated with the Sc source [27], and nitrogen vacancy-induced change in junction barrier height [28] have all been proposed. In addition, deep-level defects [29,30], impurity segregation [30], and interface roughness [31] are also known to contribute to leakage current. Thus, measurements of structural characteristics are commonly used as proxies for electrical properties.

In this study, we demonstrate low leakage current epitaxial $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films grown on GaN-templated Al_2O_3 substrates. The polarization–electric field hysteresis loops are fully saturated, and the leakage currents of our $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films grown on the GaN templates are 0.07 and 0.15 A cm^{-2} at the coercive field for 250- and 125-nm thick films, respectively. These values are one to two orders of magnitude lower than previously reported values on similar thickness films (1–19 A cm^{-2}) [21–25]. Based on the corresponding structure and property comparison, we show that crystallographic mosaicity and associated dislocation density are not the dominant parameters that control the leakage current in (Al, Sc)N/GaN stacks. Instead, we identified that the conduction mechanism of our $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ /GaN is limited by thermionic emission across the interfaces, suggesting that the density of charge-trapping defects in our films is quite low. This is distinct from previous reports of films with larger leakage currents dominated by trap-assisted Poole-Frenkel emission [24,32]. To further investigate this mechanism, an $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ film was codeposited on a lattice-matched $\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ buffer, and this film shows reduced mosaicity but increased roughness, resulting in a larger leakage current than the film on GaN buffers. This supports the conclusion that interface quality is a better predictor than crystal quality for the leakage current in heteroepitaxial (Al, Sc)N. We conclude that decreasing the density of charge-trapping point defects (e.g., nitrogen vacancies and impurities) and increasing the interface quality (decreasing surface roughness and increasing energy barrier) is effective for reducing leakage current and improving the reliability of ferroelectric nitride devices.

II. RESULTS

The crystallographic structure of the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ film deposited on the GaN template was analyzed using x-ray diffraction (XRD). Figure 1(a) shows the θ – 2θ scan

profile of the film, and Fig. 1(b) shows the ω rocking curve for the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ (0002)-diffraction peak. Besides the diffraction peaks associated with the substrate and template, only the (0002)-wurtzite peak is observed, indicating that the film is out-of-plane textured. The full width at half maximum (FWHM) of the rocking curve is 1.18° for $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ film, comparable with values reported for other films deposited by sputtering [22]. The rocking curve of off-axis (10 $\bar{1}$ 2) diffraction, which is sensitive to edge- and mixed-type threading dislocations (Fig. S1 in the Supplemental Material [33]), is comparable with the rocking curve of the surface normal-(0002) diffraction, which is sensitive to screw-type threading dislocations. The epitaxial relationship is identified using XRD φ scans around the off-axis (10 $\bar{1}$ 3)-diffraction peaks [Fig. 1(c)]. The film diffraction peak is aligned with the peak from the underlying GaN, indicating a hexagon-on-hexagon epitaxial relationship [Fig. 1(d)]. The GaN template is rotated 30° with respect to the sapphire substrates, as expected from the wurtzite/sapphire epitaxial growth mechanism [34]. The wurtzite lattice parameters, as shown in Fig. 1(d) ($a = 3.22 \text{ \AA}$ and $c = 5.03 \text{ \AA}$), are obtained from reciprocal space maps (Fig. S2 in the Supplemental Material [33]) and show that the films are fully relaxed, accommodating the 1% in-plane lattice mismatch between the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ film and GaN template.

The microstructure of the sample was investigated using scanning transmission microscopy (STEM). Differential phase contrast STEM (see Appendix B), which is sensitive to small changes in sample orientation [35], was used to highlight relative misorientation between neighboring crystallites. These images are shown in Fig. 2(a). The $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ film shows color contrast associated with crystal mosaicity. The sharp chemical interface and wurtzite phase are confirmed with energy-dispersive x-ray spectroscopy and selected area electron diffraction, respectively (Fig. S3 in the Supplemental Material [33]). The similar densities of the screw and edge dislocations can qualitatively be seen in the STEM dark field images (Fig. S4 in the Supplemental Material [33]), consistent with the XRD measurement results (Figs. 1 and S1 in the Supplemental Material [33]).

Atomic resolution STEM imaging was also employed to examine the film-substrate interface; these images are shown in Figs. 2(b) and 2(c). The annular dark field (ADF) images [Fig. 2(b)] show strong atomic number (Z) contrast between the relatively lighter elements present in the film and the heavier elements present in the substrate and, as such, highlights the location of the interface. Along with the ADF images, simultaneously acquired dDPC images [Fig. 2(c)] can be used to visualize both light and heavy elements.

Nested polarization–electric field loops, taken with a 1-kHz triangular applied electric field, show well-saturated hysteresis evidenced by their square shape [Fig. 3(a)].

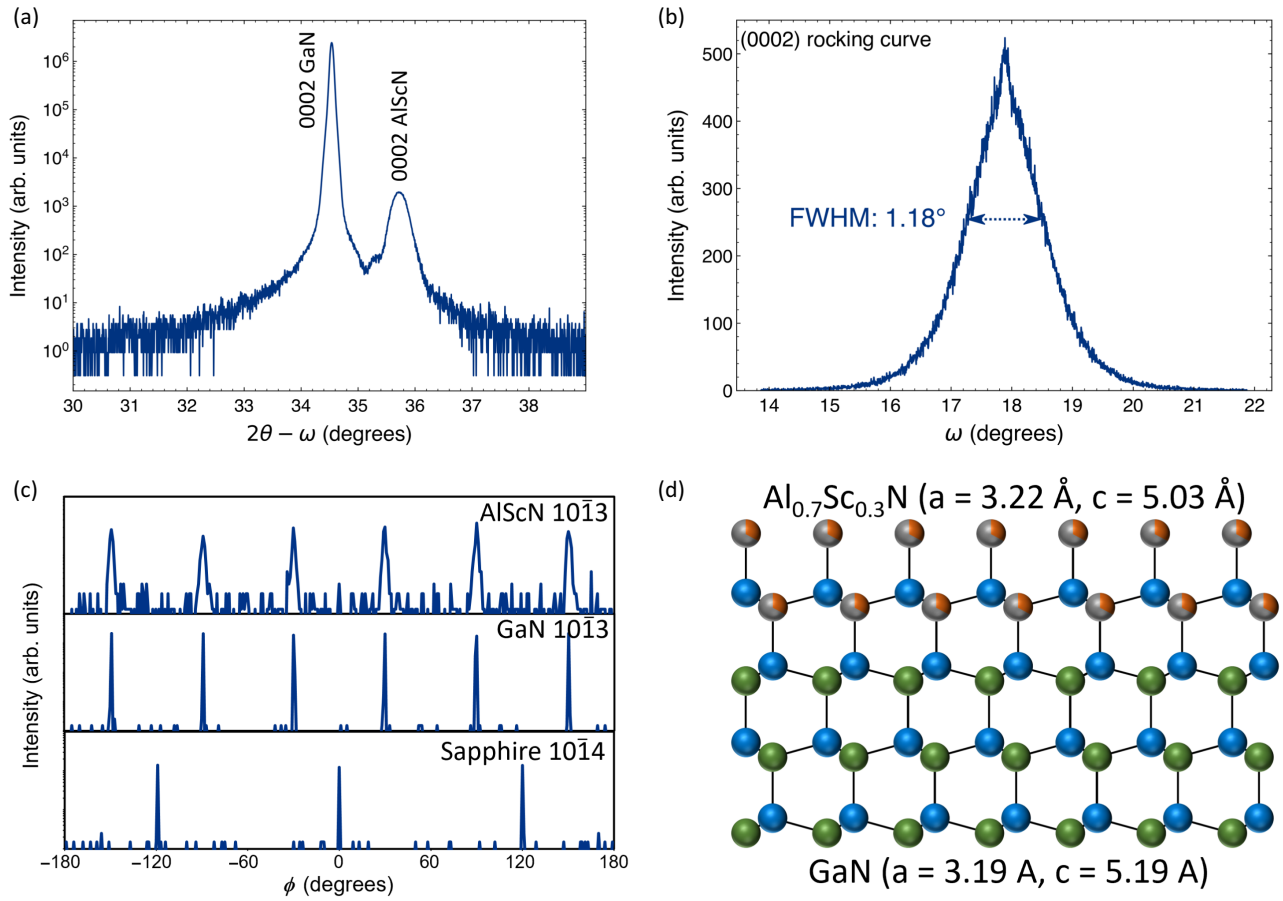


FIG. 1. XRD crystallographic analysis of $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films on GaN-templated sapphire substrates. (a) $2\theta-\omega$ profiles and (b) omega rocking curves showing (0002)-textured $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ wurtzite phase. (c) Off-axis diffraction XRD ϕ scan and (d) epitaxial relationship for $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films on GaN film in the coherent approximation, which shows that the film and template are in a hexagon-on-hexagon relationship.

The leakage current is directly measured with the current–electric field loop [Fig. 3(b)] extracted from the polarization–electric field loop under the triangular field with the maximum amplitude ($\pm 5600 \text{ kV cm}^{-1}$). To observe leakage current contributions clearly, the capacitive current density, J_c , is subtracted in the plot (see Appendix C). Besides the leakage current contribution, the sharp current peaks associated with ferroelectric switching at the coercive field are observed. Note that the current densities in Fig. 3(b) are absolute values to plot on a semilogarithmic graph.

The leakage current at the coercive field directly correlates to energy loss and charge injection during switching for ferroelectric device applications. Based on Fig. 3(b), the leakage current densities at the positive and negative respective coercive fields (i.e., 0.13 A cm^{-2} and 0.02 A cm^{-2} , respectively) are much smaller compared with the polarization current ($> 10\text{--}20 \text{ A cm}^{-1}$). The leakage current of 0.01 A cm^{-2} observed at electric fields $< 4000 \text{ kV cm}^{-1}$ is at the noise floor of the measurement equipment.

A thinner film (of 125 nm) was deposited using identical growth conditions, but half the deposition time was also investigated because thickness also affects leakage current [36,37]. The ferroelectric hysteresis loop is well-saturated, and the leakage current densities at the positive and negative coercive fields are 0.20 A cm^{-2} and 0.11 A cm^{-2} , respectively (Fig. S5 in the Supplemental Material [33]). These slightly larger leakage current densities, compared with the 250 nm film, are in good agreement with previously reported trends of increased leakage with reduced thickness [36] but still orders of magnitude lower than the reported leakage current of (Al, Sc)N with various thicknesses (i.e., 100–300 nm) [21,22,25].

To correlate the low leakage current with epitaxial crystalline quality, we grew $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films on fully relaxed $\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ buffer layers to obtain a better lattice match [Fig. 4(a)]. The $\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ film was grown on an identical GaN template using molecular beam epitaxy. It is important to note that the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films in this study, both on a GaN template and an InGaN buffer, were codeposited together

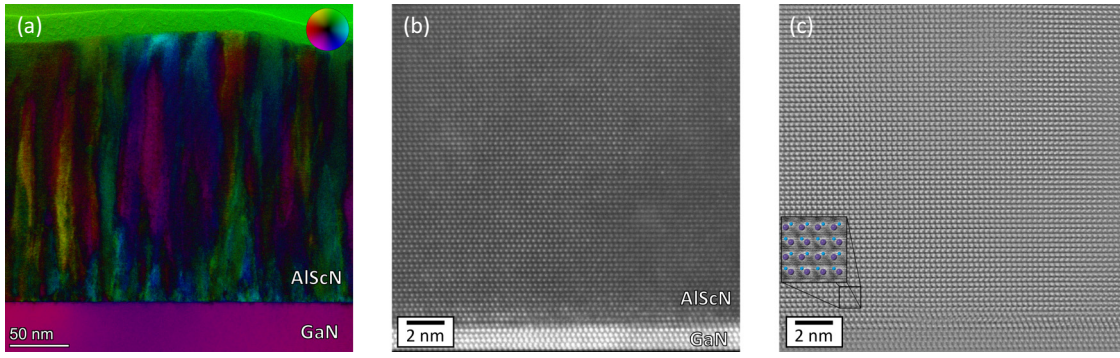


FIG. 2. STEM images and analysis of the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films grown on GaN. (a) Differential phase contrast images highlighting the relative misorientation between crystallites for the film. (b) Atomic resolution ADF images highlighting the film/template interface. (c) dDPC images acquired simultaneously with the respective ADF images.

(side-by-side in the chamber) to eliminate batch-to-batch variability. As expected from the better lattice matching, the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ stack exhibits a narrower rocking curve FWHM ($= 0.94^\circ$) and larger grain size [Figs. 4(b), 4(c) and S6 in the Supplemental Material [33]]. However, the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ film possesses a significantly rougher interface (1.92 ± 0.40 nm) than the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$ film (0.41 ± 0.19 nm), as seen in Fig. 4(d) [compared with Fig. 2(b)]. This roughness is due to limitations in the $\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ sample growth and the nitrogen-rich growth is used to prevent GaN-InN phase separation; however, it also leads to roughened surfaces. Thus, in addition to providing a better lattice match, the $\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ buffer introduces another uncontrolled variable (i.e., roughness).

Nested polarization–electric field loops, taken with a 10-kHz triangular applied electric field, show hysteresis associated with ferroelectric switching [Fig. 4(e)]. The current–electric field loop shows that the leakage current densities at the positive and negative coercive fields are 4.7 A cm^{-2} and 0.6 A cm^{-2} , respectively, which are an order of magnitude larger than the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ film on

GaN, as shown in Fig. 3(b), yet still comparable with previously reported (Al, Sc)N leakage currents. This higher leakage current leads to rounding (i.e., a lack of saturation) in the polarization–electric field hysteresis loops. Despite the improvement of the crystal mosaicity, the leakage current of the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ film is an order of magnitude larger than that of $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$ films, implying that crystal mosaicity is not a dominant parameter for leakage current.

To investigate the origins of the leakage current in these epitaxial $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films further, dc I – V characteristics are analyzed, as shown in Fig. 5. Both 250-nm $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$ and $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ films show linear conduction for electric fields $< 1000 \text{ kV cm}^{-1}$ and an exponential current increase at higher fields [Fig. 5(a)]. The leakage current density of $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ is higher than that of $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$, which is in good agreement with the polarization and current loops shown in Figs. 3 and 4. This exponential behavior can be correlated with the dominant conduction mechanism [38]. Conduction in the (Al, Sc)N films has been previously reported to be limited by either

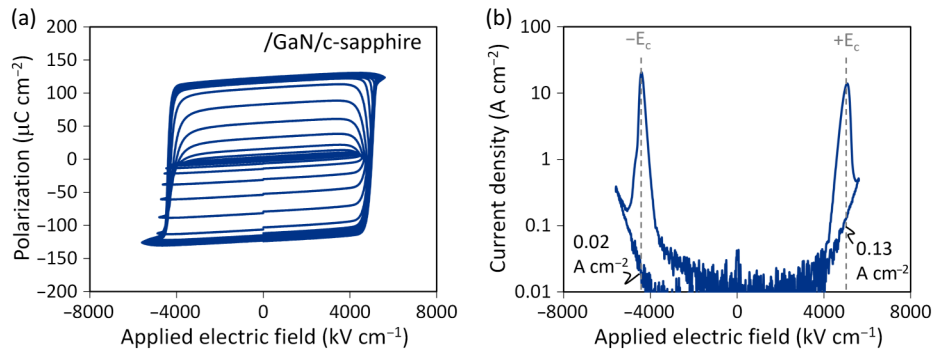


FIG. 3. Ferroelectric properties characterization for $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films on GaN-templated substrates. (a) Nested ferroelectric hysteresis loops showing the polarization saturation. The excitation triangular field frequency is 1 kHz. (b) Current loops, subtracting out the capacitive current density, showing record low leakage currents along with the polarization switching current spike.

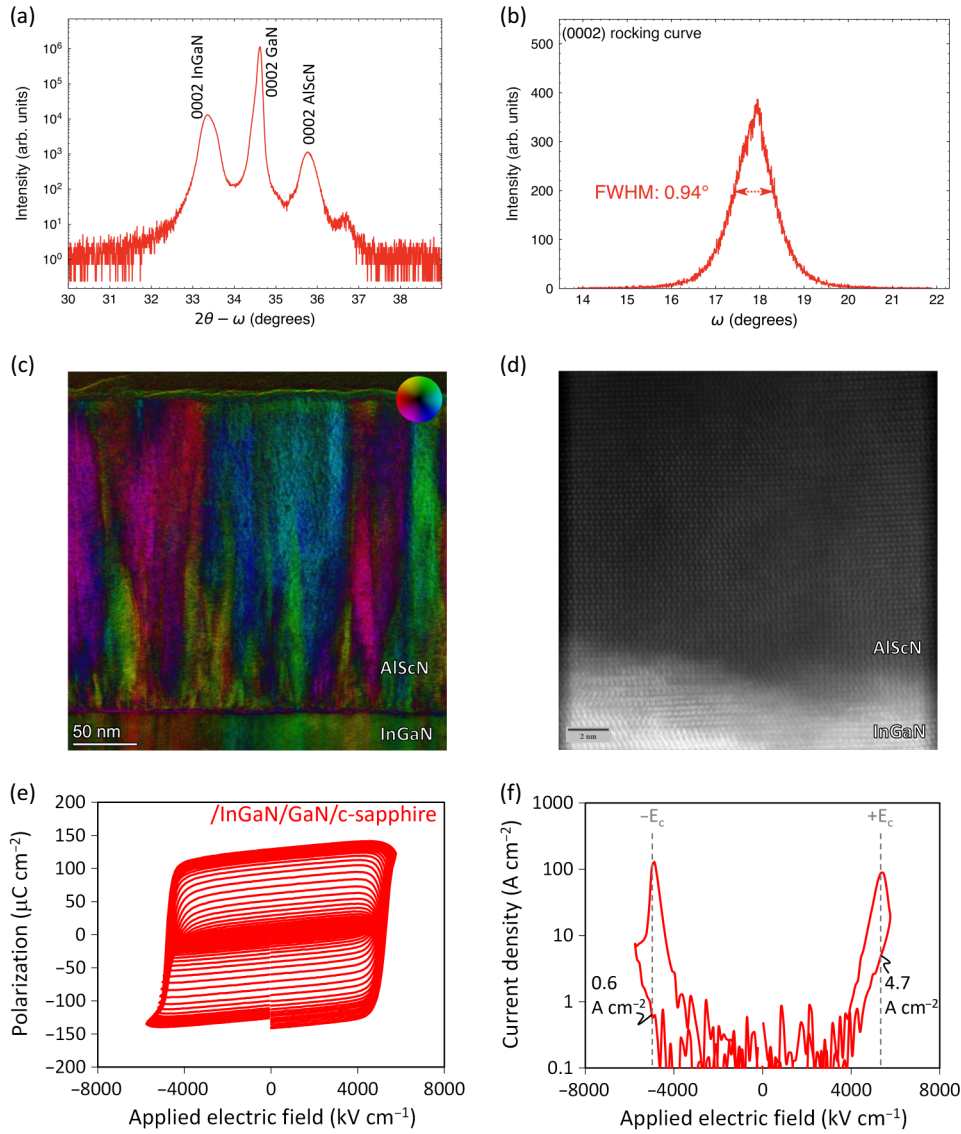


FIG. 4. Characterization of $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films on lattice-matched $\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ templated substrates. (a) 2θ - ω profiles and (b) omega rocking curve scans showing (0002)-textured $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ wurtzite phase. (c) Differential phase contrast image highlighting the relative misorientation between crystallites for the film. (d) Atomic resolution ADF images highlighting the film/template interface. (e) Nested ferroelectric hysteresis loops with an excitation triangular field frequency at 10 kHz. (f) Current loops showing switching current and leakage current.

thermionic emission [28] or Poole-Frenkel trap-assisted transport [24,32].

To identify the conduction mechanism of our (Al, Sc)N films for electric fields $>1000 \text{ kV cm}^{-1}$, the slope of the logarithm of the current density- $E^{1/2}$ curve in the nonlinear emission regime is examined, as shown in Fig. 5(b). The fitted slopes are 6.7×10^{-4} and $7.5 \times 10^{-4} \text{ m}^{1/2} \text{ V}^{-1/2}$ for $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$ and $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$, respectively. The slope is compared with the thermionic emission models, as expressed in Appendix C [39–41]. With the permittivity determined by the refractive index providing the high-frequency permittivity [42], $\epsilon_r = 4.7$ for $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ [43],

the calculated slope from the thermionic emission model is $6.8 \times 10^{-4} \text{ m}^{1/2} \text{ V}^{-1/2}$, which is in good agreement with the experimental results.

The slopes of the logarithms of $J/E-E^{1/2}$ curves for electric fields $>1000 \text{ kV cm}^{-1}$ do not fit the calculated slope based on the Poole-Frenkel emission model [Fig. 5(c)]. The experimental slopes are $5.0 \times 10^{-4} \text{ m}^{1/2} \text{ V}^{-1/2}$ and $5.7 \times 10^{-4} \text{ m}^{1/2} \text{ V}^{-1/2}$ for $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$ and $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$, respectively, which are less than half of the calculated value of $1.36 \times 10^{-3} \text{ m}^{1/2} \text{ V}^{-1/2}$. Thus, we conclude that the dominant conduction mechanism at $E > 1000 \text{ kV cm}^{-1}$ is thermionic emission for both the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$

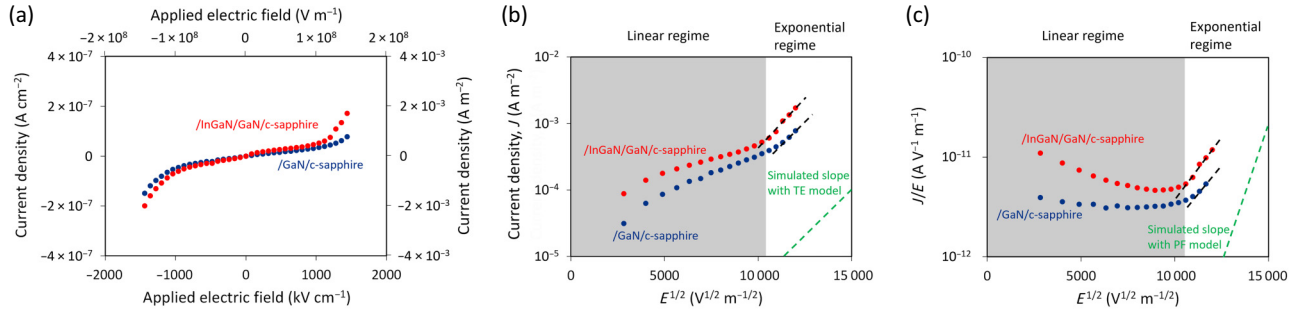


FIG. 5. Analysis of the leakage current mechanisms for the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$ and $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ films. (a) Current density–electric field curves showing linear conduction at $<1000 \text{ kV cm}^{-1}$ and exponential conduction at $>1000 \text{ kV cm}^{-1}$. (b) $\text{Log}[J]-E^{1/2}$ plot to compare the slope of the exponential conduction regime to the thermionic emission model. (c) $\text{Log}[J/E]-E^{1/2}$ plot to compare the slope of the exponential conduction regime to the Poole-Frenkel (PF) emission model. We find agreement between the data and the thermionic emission model but not with the Poole-Frenkel model.

and $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ films deposited in this study.

III. DISCUSSION

Both the 250- and 125-nm films on the GaN template studied here showed significantly lower leakage current than that of previously reported (Al,Sc)N/GaN films. Table I compares the properties of ferroelectric (Al,Sc)N/GaN films reported to date, including this study. The films on the GaN template in this study show one to

two orders of magnitude lower leakage current than the other reported films. The leakage current density, listed in Table I, is an average value of the current under $+E_c$ and $-E_c$. The asymmetry is attributed to two convoluted factors: asymmetric leakage current due to the different electrode contacts (n-GaN or Pt) and asymmetric E_c because of trapped charges [44–46] and/or polarity dependence on wake-up behavior [47]. The low leakage current of $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$ films in this study provides a meaningful step toward the integration of sputtered (Al,Sc)N ferroelectrics in microelectronics devices.

TABLE I. Property comparison of epitaxial (Al,Sc)N film on GaN templates. Leakage currents are given as the average between positive and negative switching leakage.

	This study		Schönweger [22]		Wang [21]	Wang [23]	Wang [24]	Wolff [25]
Sc content x	0.3		0.28		0.20	0.21	0.18	0.15
Thickness	250 nm	125 nm	300 nm	100 nm	100 nm	100 nm	100 nm	230 nm
Template	n-GaN	n-GaN	n-GaN	n-GaN	n-GaN	n-GaN	n-GaN	n-GaN
E_c	4.7 MV cm^{-1} @ 1 kHz	5.1 MV cm^{-1} @ 1 kHz	4.6 MV cm^{-1} @ 1.5 kHz	5.5 MV cm^{-1} @ 2 kHz	4.2 MV cm^{-1} @ 10 kHz	4.6 MV cm^{-1} @ 10 kHz	$\sim 7 \text{ MV cm}^{-1}$ @ 40 kHz	5.5 MV cm^{-1} @ 1.5 kHz
J at E_c with ac measurement	0.07 A cm^{-2}	0.15 A cm^{-2}	$\sim 1 \text{ A cm}^{-2}$	$\sim 4 \text{ A cm}^{-2}$	$\sim 7 \text{ A cm}^{-2}$	$\sim 19 \text{ A cm}^{-2}$	$\sim 1 \text{ A cm}^{-2}$	$\sim 8 \text{ A cm}^{-2}$
J with dc measurement	$7.8 \times 10^{-8} \text{ A cm}^{-2}$ @ 1.4 MV cm^{-1}	$8.5 \times 10^{-8} \text{ A cm}^{-2}$ @ 1.4 MV cm^{-1}	—	—	—	$\sim 1.5 \times 10^{-5} \text{ A cm}^{-2}$ @ 1.4 MV cm^{-1}	$\sim 3 \times 10^{-5} \text{ A cm}^{-2}$ @ 2.5 MV cm^{-1}	—
Device yield	8/10	7/10	—	—	—	—	—	—
(0002) rocking curve FWHM	1.18°	1.27°	$\sim 1^\circ$	$\sim 1^\circ$	—	$\sim 0.4^\circ$	$\sim 0.05^\circ$	$\sim 0.07^\circ$
Device size	50 μm in diameter		100 $\mu\text{m} \times 100 \mu\text{m}$	50 $\mu\text{m} \times 50 \mu\text{m}$	20 μm in diameter	20 μm in diameter	20 μm in diameter	20 μm in diameter
Deposition technique	Rf sputtering		Pulsed dc sputtering		MBE			MOCVD

Rocking curve FWHM is a reliable proxy measurement for structural quality because it is sensitive to dislocation densities [48] and provides a convenient comparison of the structural quality across films from different studies. The rocking curve FWHM of the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films is comparable with earlier-reported sputter deposited (Al, Sc)N/GaN [22] and wider than molecular beam epitaxy (MBE) or metalorganic chemical vapor deposition (MOCVD) grown films [23–25]. This comparison clearly shows that epitaxial structural quality is not the dominant determinant for leakage current, which is consistent with the comparison between the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$ and $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ films in this study.

Our observed leakage current mechanism, coupled with the comparison between $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$ and $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ films, provides further evidence of the origin of leakage current in (Al, Sc)N ferroelectric devices. The mechanism that limits conduction in our $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films is thermionic emission at the interface, which is distinct from the reported studies showing only Poole-Frenkel trap-assisted transport in (Al, Sc)N [24,32]. These studies presented large leakage current densities at the coercive fields (i.e., $\sim 3 \text{ A cm}^{-2}$ and $\sim 30 \text{ A cm}^{-2}$) and rounded hysteresis loops. Because Poole-Frenkel emission is trap-mediated carrier conduction in the bulk [39], the low leakage current of our $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$ is attributed to the low trap densities in the bulk, likely associated with the low defect density. This low trap density in the bulk enables the observation of the thermionic emission regime in the high electric field range in our epitaxial $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films. Dislocations associated with crystallographic mosaicity appear to be inactive as a trap state that causes Poole-Frenkel transport. Hence, crystallographic mosaicity is not critical for the leakage current, as discussed above. It is noteworthy that there is no other significant difference between our films and the previously reported films [21,22], including the grain size and the impurity segregation based on the TEM observation (Fig. 2). Thus, the Poole-Frenkel traps-assisted transport in these literature studies are likely associated with point defects, such as nitrogen vacancies and/or impurities, which is consistent with other AlN-based semiconductor materials [49–51].

The larger thermionic emission current in our $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ film compared with the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$ film can be attributed to the interface roughness and associated average electric field increase; namely, the horizontal shift of the $\log[J]-E^{1/2}$ curve in Fig. 5(b). It has been analytically proven that the average electric field in an insulator increases up to 15% due to interface roughness [31]. Our experimental results, which show a shift of the effective electric field where thermionic emission becomes dominant [$\sim 1200 \text{ kV cm}^{-1}$ for $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$ and $\sim 1000 \text{ kV cm}^{-1}$ for $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$, Fig. 5(a)], are in good agreement with the calculated roughness

contribution. This $\sim 20\%$ increase in the average electric field is substantial for the leakage current that exponentially increases with respect to the electric field. Therefore, we conclude that interface roughness leads to the leakage current increase of the sputtered $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ film compared with the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$ film despite the film mosaicity improvement (FWHM = 0.94° compared with FWHM = 1.18°) and associated dislocation density decrease. As discussed above, structural dislocations are likely inactive as carrier trap states causing Poole-Frenkel transport, which implies that reduced dislocation density does not significantly improve the leakage current.

IV. CONCLUSION

We have demonstrated epitaxial $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}/\text{GaN}$ thin film deposited via rf reactive magnetron sputtering on sapphire substrates exhibiting low leakage current, i.e., 0.07 A cm^{-2} , which is one to two orders of magnitude lower than previously reported for epitaxial (Al, Sc)N/GaN stacks. The x-ray rocking curve FWHM is 1.18° , which is comparable with those reported for sputtered (Al, Sc)N films with $\sim 10\times$ larger leakage currents, and three times wider than MBE or MOCVD deposited films that show $\sim 100\times$ larger leakage currents. This indicates the crystal quality and associated dislocation density of the (Al, Sc)N film is not the dominant factor determining the leakage current. Instead, the conduction mechanism limiting the leakage current is identified as thermionic emission, which is defined by the interface carrier injection and is influenced by surface roughness more than the bulk structural quality. In support of this conclusion, $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ grown on lattice-matched $\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ buffer layers were observed to have improved structural properties and identical current conduction mechanisms while exhibiting an order of magnitude higher leakage current, which is attributed to increased interface roughness and associated effective electric field increase. These results demonstrate progress in mitigating the leakage current of (Al, Sc)N/GaN heteroepitaxial nitride ferroelectrics stacks, and show the importance of increasing surface and interface quality for ferroelectric devices with improved memory retention for use in energy-efficient microelectronics.

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The authors declare no conflict of interest.

DATA AVAILABILITY

The data affiliated with this study are available from the corresponding author upon reasonable request.

APPENDIX A: SAMPLE PREPARATION

The 250-nm $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ films were deposited on an (0001)-oriented GaN-on-sapphire template and MBE deposited (In, Ga)N buffer layer via reactive rf magnetron sputtering using the following growth conditions: 2 mTorr of Ar/N_2 (15/5 sccm flow) and a target power density of $6.6 \text{ W}/\text{cm}^2$ on a 2" diameter $\text{Al}_{0.7}\text{Sc}_{0.3}$ alloy target (Stanford Advanced Materials). The substrate was rotated and heated to 400°C during deposition. The base pressure, partial oxygen, and water vapor pressure at 400°C were $<2 \times 10^{-7}$ torr, $P_{\text{O}_2} < 2 \times 10^{-8}$ torr, and $P_{\text{H}_2\text{O}} < 1 \times 10^{-7}$ torr, respectively. The top Au (100 nm)/Ti (20 nm) contacts, which are 50 μm in diameter, were deposited on the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ film via electron beam evaporation through a photolithographic pattern.

The InGaN buffer layer was grown in a Riber compact 21 molecular beam epitaxy (MBE) system using previously published methods [52]. An (0001)-oriented GaN-on-sapphire template (6- μm thick, Si-doped, 3LCorp) was used as a substrate. The GaN surface was first regrown using metal-modulated epitaxy, monitoring the III/V ratio by reflection high energy electron diffraction (RHEED) intensity transients [53,54]. The GaN regrowth was $\sim 130 \text{ nm}$ in thickness, after which the substrate thermocouple setpoint was lowered to 600°C , and the III-V ratio was lowered to a slightly nitrogen-rich regime (III/V = 0.92) targeting 18% In. The InGaN was grown to a total thickness of 100 nm. During growth, the relaxed in-plane lattice constant was monitored by RHEED, and the In/Ga ratio was adjusted after relaxation to obtain an in-plane lattice constant of approximately 3.23 Å.

APPENDIX B: STRUCTURAL ANALYSIS VIA XRD AND TEM

The crystal structure was analyzed using a Rigaku Smartlab with Cu $K\alpha$ radiation monochromated by a

2-bounce Ge (220) channel cut crystal and columnated with a parabolic mirror. (0002) rocking curves, used to analyze mosaic tilt, were obtained using a slit of 0.5 mm on the diffracted beam path. (10 $\bar{1}2$) rocking curves, used to analyze combined mosaic tilt and twist, were obtained in a skew-symmetric geometry by tilting χ using a slit of 1 mm on the diverging beam path [55].

Scanning transmission electron microscopy (STEM) was conducted on a Thermo Fisher Themis 200 G3 aberration-corrected microscope operating at 200 keV, with a probe semiangle of convergence of 17.9 mrad. Differential phase contrast [56] (DPC) imaging was conducted using a four-segment annular detector, with a camera length corresponding to inner and outer collection angles of 11 and 43 mrad, respectively. Annular dark field (ADF) images (46–200 mrad inner and outer collection angles) were collected simultaneously with DPC. The roughness of the film-substrate interfaces was measured by marking the position of the interface across the width of each DPC image and then calculating the root-mean-squared deviation of the interface height from its average within each image. Roughness values measured by this method may not be comparable with the roughness values measured with techniques owing to the projection of one of the interface dimensions in the STEM images, but they are comparable with each other.

APPENDIX C: ELECTRICAL CHARACTERIZATION

Ferroelectric polarization–electric field hysteresis measurements and current–electric field loop measurements were taken with a Precision Multiferroic system from Radiant Technologies. The applied triangle excitation field was up to 5.6 MV cm^{-1} at 1 kHz for the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ on the GaN template, and 5.8 MV cm^{-1} at 10 kHz for the $\text{Al}_{0.7}\text{Sc}_{0.3}\text{N}$ on the $\text{In}_{0.18}\text{Ga}_{0.82}\text{N}$ template.

The capacitive current was subtracted to extract the leakage current contribution from the current–electric field loop. The capacitive current is defined as

$$j_c = \frac{1}{A} \frac{dQ}{dt} = \frac{CdV}{Adt} = \frac{\epsilon_r \epsilon_0}{d} \frac{dV}{dt},$$

where A is the device area, C is the capacitance, ϵ_r is the relative permittivity, ϵ_0 is the permittivity of free space, and d is the film thickness. This constant current in the triangular excitation (dV/dt is constant) was subtracted from the raw current loop data.

Steady-state I – V curves were taken with Keithley 4200A-SCS. A step voltage starting from 0 to $\pm 36 \text{ V}$ (1440 kV cm^{-1}) with a step of 2 V (a delay time of 5 s at each step) was applied to obtain a steady-state current flow at each voltage step.

The thermionic emission model is expressed as [39–41]

$$J = RT^2 \exp \left[-\frac{\phi_s - (1/2)(q^3/(\pi \epsilon_r \epsilon_0))^{1/2} E^{1/2}}{kT} \right],$$

where R is the Richardson constant, T is the temperature, ϕ_s is the metal-insulator barrier height, q is the unit of the electron charge, ϵ_r is the relative permittivity at high frequency [42], and ϵ_0 is the permittivity at vacuum.

The Poole-Frenkel emission model is expressed as [39–41]

$$J = CE \exp \left[-\frac{\phi_{PF} - (q^3/(\pi \epsilon_r \epsilon_0))^{1/2} E^{1/2}}{kT} \right],$$

where C is a constant and ϕ_{PF} is the ionization potential of the Poole-Frenkel centers.

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