

RESEARCH ARTICLE

Low Temperature Growth of High-Quality Wurtzite Ferroelectric Through Quasi-van der Waals Epitaxy

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ABSTRACT

Wurtzite-type aluminum scandium nitride (AlScN) has emerged as a potential candidate for next-generation ferroelectrics due to large polarization, high Curie temperature and compatibility with semiconductor technology. However, strategies that enable the low-temperature growth of high-quality films, and the exfoliation of freestanding membranes are still lacking, largely hindering integration in functional devices. Here, we demonstrate the growth of high-quality AlScN films at the low temperature of 150°C on substrates with distinct crystal symmetries, achieved by using a two-dimensional transition metal dichalcogenide (TMDC) monolayer as a buffer. The high crystallinity is stemmed from the quasi-van der Waals epitaxy, which is universally demonstrated on a wide range of TMDC materials. The TMDC buffers further enable the fabrication of freestanding AlScN membranes, exhibiting excellent ferroelectric properties including high remanent polarization, low leakage current and rapid switching kinetics. Our findings provide a low-thermal-budget approach with high integration flexibility to develop wurtzite-based ferroelectric, piezoelectric, and electro-optic devices.

1 | Introduction

Ferroelectric thin films with nonvolatile and switchable polarization play a crucial role in nonvolatile memory, in-memory computing, electro-optic modulator and electro-mechanical device [1–5]. Aluminum scandium nitride (AlScN), a recently emerged ferroelectric, exhibits robust ferroelectric phase, high remanent polarization, high Curie temperature and good compatibility with complementary metal oxide semiconductor (CMOS) architecture, rendering it as a strong candidate for next-generation

ferroelectric devices [6–9]. To fully exploit this material, it is highly demanded to grow AlScN films with high crystalline quality (for robust ferroelectricity), at low temperature (for CMOS compatibility), and on arbitrary substrates (for scalable hetero-integration), which are challenging to achieve simultaneously [2, 3]. Epitaxial AlScN films have been grown on isostructural single-crystal substrates (e.g., SiC) or thick buffers (e.g., GaN or AlN on Al₂O₃) by various physical and chemical vapor deposition techniques [10–15]. However, these methods rely on using specialized single-crystal substrates or buffers with deposition tem-

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peratures exceeding 400°C, making it incompatible with CMOS back-end-of-line standards. For integration on substrates with dissimilar structures, magnetron sputtering offers a low-thermal-budget alternative by using thick buffers such as (111)-oriented Pt, TiN, or HfN_{0.4} [6, 16–18]. Recent developments of high-power impulse magnetron sputtering further improve the crystallinity of thick AlScN films [19–22]. Unfortunately, the dissimilar crystal symmetry and lattice mismatch between AlScN and buffers still degrade the crystallinity of ultrathin films due to excessive defects near interface [6, 16, 17, 23–32]. Moreover, the strong chemical bonds prevent the exfoliation of AlScN freestanding membranes, which has received increasing attentions for van der Waals (vdW) devices and flexible electronics [33–36]. To date, a universal strategy that enables simultaneous low-temperature high-quality growth, integration on arbitrary substrates, and freestanding exfoliation has yet to be developed for AlScN.

The discovery of vdW epitaxy based on vdW interaction between two-dimensional (2D) materials and deposited adatoms, as well as quasi-vdW epitaxy that involves chemical bond formation, offers a promising approach to solve the above challenges [34, 37–40]. Transition metal dichalcogenides (TMDCs), such as MoS₂ and WS₂, possess the hexagonal structures with an in-plane lattice constant ranging from 3.18 to 3.29 Å, exhibiting the similar in-plane symmetry and lattice constant to the III-nitride compounds (e.g., $a = 3.22$ Å for Al_{1-x}Sc_xN with $x = 0.26$) [3, 41]. Therefore, they could potentially serve as excellent templates for the growth of III-nitrides, as supported by recent reports on growing binary GaN and AlN films on TMDCs materials [42, 43]. The large-scale growth and transfer of TMDC monolayers have been demonstrated on various substrates, offering the pathway to overcome the substrate limitations. Moreover, the vdW nature of TMDCs enables mechanical exfoliation, offering potential for fabricating freestanding AlScN membrane. These features render the (quasi-)vdW epitaxy as a promising strategy for the synthesis of high-quality ferroelectric AlScN films, which, however, has not yet been explored [42, 43].

In this study, we demonstrate the low-temperature growth of AlScN films with high crystalline quality and excellent ferroelectric properties, using 2D TMDC as the buffer as illustrated in Figure 1. Highly textured Al_{0.74}Sc_{0.26}N films with *c*-axis orientation and large grain size were synthesized by using conventional magnetron sputtering at 150°C. Using integrated differential phase contrast scanning transmission electron microscopy (iDPC-STEM) and theoretical calculations, we reveal a quasi-vdW epitaxial growth mode, wherein AlScN chemically bonds to the interfacial TMDC monolayer while the rest TMDC maintains the vdW interactions. The universality of this growth methodology is further demonstrated for a wide range of 2D TMDC materials. Moreover, high-quality AlScN films were also successfully deposited on Si substrates by using various TMDCs with monolayer thickness (~0.5 nm), highlighting the potential for integration on arbitrary substrates. This strategy further enables mechanical exfoliation and transfer of freestanding AlScN membranes, which exhibit excellent ferroelectric properties including high polarization, low leakage current and rapid switching kinetics. Our findings open a new avenue for implementing AlScN in ferroelectric, piezoelectric, and optoelectronic devices.

2 | Results and Discussion

2.1 | Growth of AlScN on MoS₂ Buffers

To investigate the feasibility of using 2D TMDCs for AlScN growth, we first used MoS₂ with a varying number of layers as the buffer. AlScN films were grown via radio frequency (RF) dual-target co-sputtering using Al and Al_{0.57}Sc_{0.43} alloy targets (see Experimental Methods Section for details). The growth temperature was 150°C to minimize the thermal budget while maintaining high crystallinity of AlScN (see Figure S1 for further results). As shown in Figure 2a, the θ -2 θ X-ray diffraction (XRD) scans reveal strong diffraction peaks at 35.8° in all samples with MoS₂ buffers on Al₂O₃, revealing a strong (0002) textured AlScN films. In contrast, the film grown directly on Al₂O₃ substrate shows an almost negligible diffraction peak, signifying the critical role of MoS₂ buffer. This is further supported by comparing the full width of half maximum (FWHM) of the rocking curves. As shown in Figure 2b, AlScN film grown on MoS₂ buffer exhibits a significantly lower FWHM (~0.62°) as compared to that on the commonly used Pt buffer (~2.55°), revealing a largely improved crystalline quality enabled by MoS₂ [31, 44]. Further analysis of lattices reveals strain relaxation for AlScN films on both buffers (see Figures S4 and S5). Figure 2c shows the FWHM of AlScN (0002) as a function of MoS₂ layer number, confirming the growth of highly textured AlScN films on MoS₂ down to monolayer thickness (FWHM of ~1.8°). Note that the FWHM decreases as increasing MoS₂ thickness, suggesting that the AlScN growth is not likely due to the remote epitaxy mediated by electrostatic interaction between films and substrates [38]. Moreover, AlScN films with varying thicknesses were also grown on MoS₂ monolayer. The (0002) diffraction peaks are observed for AlScN down to 7 nm, suggesting the feasibility to obtain ultrathin films (see Figure S6 for further results). The FWHM decreases with increasing AlScN thickness, consistent with previous reports [17, 44, 45].

Figure 2d shows the Raman spectra of MoS₂ buffer layers after depositing AlScN. The characteristic peaks of MoS₂, i.e., E_{2g}^1 (~383 cm⁻¹) and A_{1g} (~408 cm⁻¹), are observed for the heterostructures with more than two layers of MoS₂. Moreover, the frequency of E_{2g}^1 (A_{1g}) peak decreases (increases) as increasing the layer number, consistent with the layer-number dependence of Raman spectra for MoS₂ in previous report [46]. These results indicate that the MoS₂ buffer retains its structural integrity after AlScN deposition. The absence of characteristic Raman spectra in the sample with monolayer MoS₂ could be attributed to the formation of chemical bonds at the interfacial layer, which will be discussed in the following section.

Figure 2e compares the growth of AlScN films using different methods and buffer layers (see Table S1 for detailed information). For epitaxial growth on isostructural buffers like GaN, the high crystallinity with FWHM <1° is achieved, yet all processes require growth temperature over 400°C [10–14]. On the other hand, the growth of AlScN films at relatively low temperatures via conventional DC and RF magnetron sputtering has been realized using Pt and TiN buffers, which however, usually exhibit the FWHM larger than 2° [16, 23, 28–30]. Recent advances of high-energy and ionized sputtering techniques provide a route to

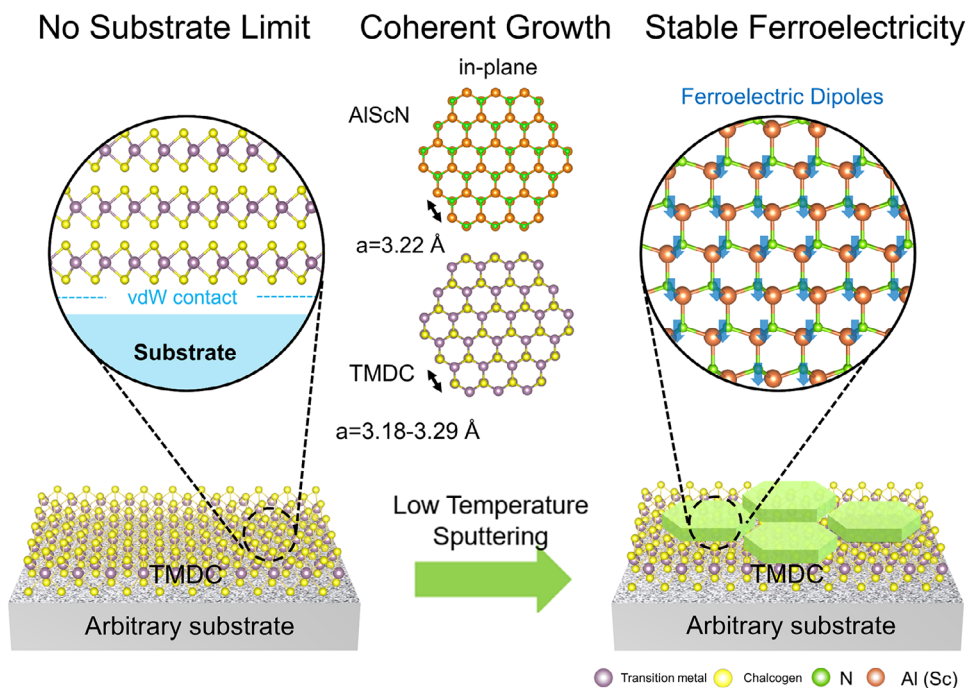


FIGURE 1 | Schematic diagram of quasi-van der Waals (vdW) epitaxy. The transition metal dichalcogenides (TMDCs), with similar crystal symmetry and lattice, serve as good templates to enable the low-temperature growth of AlScN on arbitrary substrates.

achieve low-temperature growth (from room temperature to 300°C) of thick films (>150 nm) with improved crystallinity (FWHM of $\sim 1.5^\circ$). Notably, our approach by using MoS₂ buffers enables the growth of high-quality thin films (FWHM of $\sim 0.6^\circ$) at the low temperature of 150°C by using conventional sputtering, standing out among the available growth strategies.

2.2 | Atomic-Scale Microscopy of AlScN/TMDC Heterostructure

To reveal the growth mechanism, we performed cross-sectional STEM imaging. The annular dark-field image reveals that AlScN on MoS₂ buffer exhibits the grain size of 30–50 nm (see Figure S5a), larger than that on Pt buffer (about 5–10 nm) [44, 45]. This is consistent with the lower FWHM of rocking curve as shown in Figure 2b. We further used iDPC-STEM to probe the atomic structures of both AlScN and MoS₂ layers across the interface. As shown in Figure 3a, AlScN grown on 8-layer MoS₂ exhibits high crystallinity and strong out-of-plane (0002) texture. Clear tilted dumbbell structures of Al (Sc)-N are observed within the AlScN layer (Figure 3b), revealing a uniform nitrogen-polar (N-polar) state. By using a centrosymmetric layer-hexagonal structure as a reference, the polarization is estimated to be around $131.1 \pm 3.1 \mu\text{C}/\text{cm}^2$ [47]. Figure 3c shows the atomic resolved iDPC image of the interfacial region, with the atomic structures of AlScN and MoS₂ overlaid. It is observed that the Al atoms are directly above S atoms at the interface with a spacing around 1.88 Å, much shorter than the vdW interlayer spacing of MoS₂ (around 3.32 Å, see Figure 3a). Similar interfacial structure is also shown in AlScN on a monolayer MoS₂ buffer (see Figure S7). These results suggest the formation of chemical bonds at the interface, indicating the quasi-vdW epitaxial growth of AlScN on MoS₂ [48].

To further investigate the AlScN/MoS₂ interface, we performed density functional theory (DFT) calculations. As shown in Figure 3d, several possible interfacial atomic configurations were considered: (i) metal-polar (M-polar) AlScN with N atoms on top of S atoms; (ii) N-polar AlScN with N atoms on top of S atoms; (iii) N-polar AlScN with Al atoms on top of S atoms; (iv) M-polar AlScN with Al atoms on top of S atoms. The calculation results show that the configuration iii, with chemical bonds between Al and S atoms, has the lowest formation energy among all states, consistent with the experimental observations. These results corroborate the quasi-vdW epitaxy for AlScN on MoS₂ buffer. The TMDC templates facilitate the initial growth of AlScN layer via chemical bonding due to the similar in-plane symmetry and lattice, leading to a well-aligned crystallographic orientation and large grain sizes of AlScN. Meanwhile, the vdW interactions within TMDC layers are still preserved. The growth is mainly dictated by the top TMDC layers, which therefore isolate the influence of Coulomb interaction and misfit strain of the underlying substrates [34, 38].

2.3 | Universality of Quasi-vdW Epitaxy

To elucidate the universality of this strategy, we first grew AlScN films on various TMDC materials with monolayer thickness on Al₂O₃ substrates (left panel of Figure 4a), including the CVD-grown PtSe₂ ($a = 3.70 \text{ Å}$), MoSe₂, WSe₂ ($a = 3.29 \text{ Å}$), MoS₂ and WS₂ ($a = 3.18 \text{ Å}$). Moreover, AlScN films were also grown on the monolayer graphene ($a = 2.46 \text{ Å}$) transferred onto Al₂O₃ as a reference [41]. Figure 4b shows the θ -2 θ scans of 50 nm AlScN films grown on these 2D buffers. For TMDCs with lattice constants close to AlScN, such as MoS₂, WS₂, MoSe₂, and WSe₂, a pronounced (0002) diffraction peak is observed, revealing the

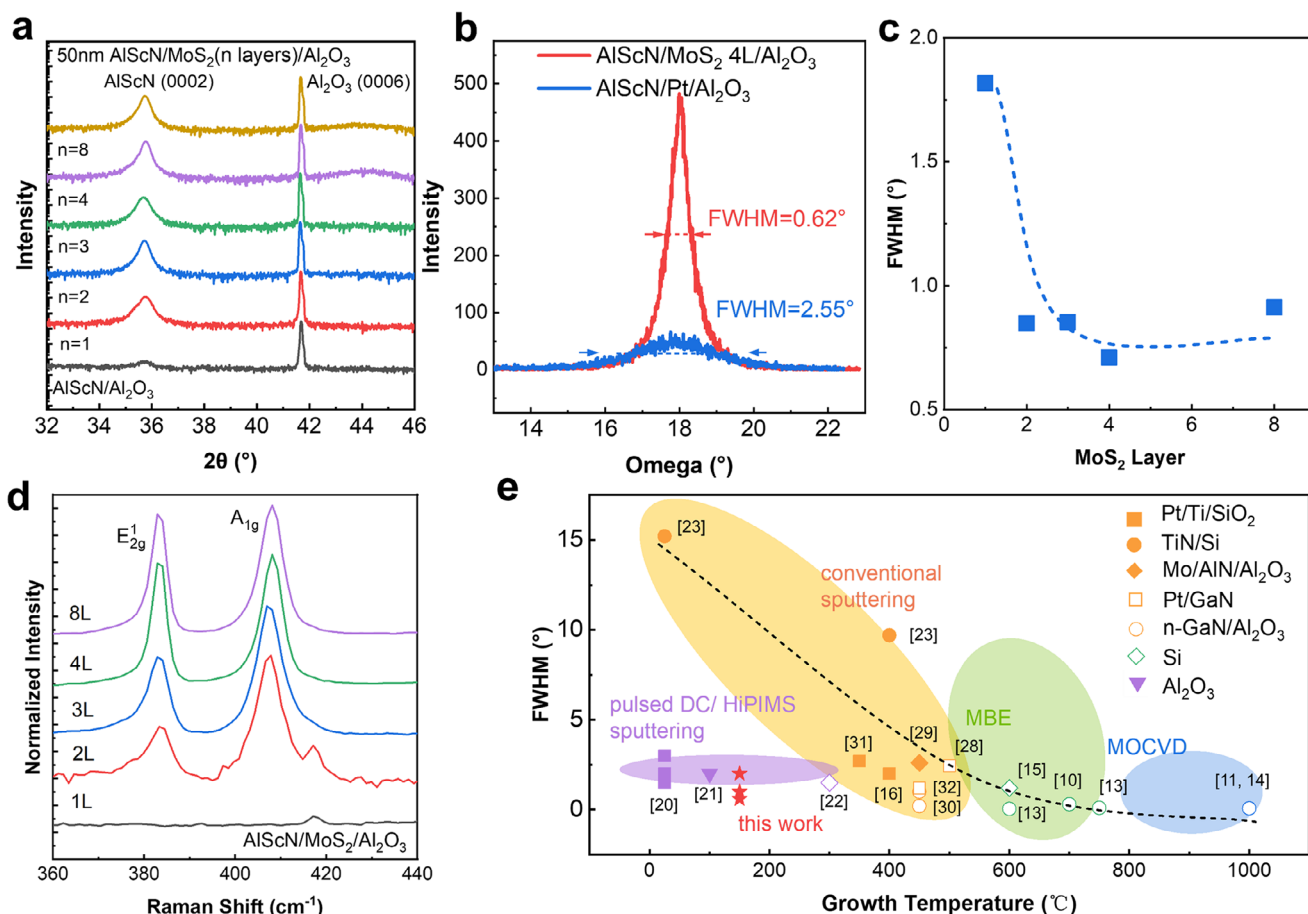


FIGURE 2 | Structural analysis of MoS₂ buffered AlScN. (a) θ - 2θ scan results of 50 nm-thick AlScN on Al₂O₃ substrates, using MoS₂ buffers with different layer numbers (n). (b) Rocking curves of 50 nm AlScN grown on Pt/Al₂O₃ and 4-layer MoS₂/Al₂O₃ at the temperature of 150°C. (c) The full width at half maximum (FWHM) of AlScN (0002) as a function of MoS₂ layer number. (d) Raman spectra of MoS₂ after AlScN growth. (e) Summary of growth temperature and FWHM of AlScN (0002) by using different substrates and buffer layers, where the different colored regions represent different synthesis techniques.

formation of high-quality AlScN films. In contrast, when the lattice of buffers is either too large (PtSe₂) or too small (graphene), the AlScN diffraction peak significantly diminishes, indicating a reduction of crystalline quality.

Next, we investigated the growth of AlScN films on the Si (001) substrate, which exhibits a distinct crystal structure and large lattice mismatch as comparing to AlScN. As schematically shown in Figure 4a (right panel), various monolayer-thick TMDCs were transferred onto Si substrates by using a polymethyl methacrylate (PMMA)-assisted process, followed by the deposition of 50 nm AlScN films via magnetron sputtering at 150°C. Figure 4c shows strong (0002) peaks for all AlScN films grown on these transferred TMDC buffers (Figure 4c), revealing the textured growth (see Figures S8 and S9 for further structural analysis). Ferroelectric measurements further verify robust ferroelectricity for these AlScN films (see Figure S10). In a sharp contrast, AlScN films deposited directly on Si substrates without TMDC buffers show a significantly reduced (0002) diffraction peak (black curve in Figure 4c), further corroborating the critical role of TMDC buffers. We note that the AlScN films grown on the transferred TMDCs show slightly larger FWHM when comparing to those on

the CVD-grown TMDCs (see Figure S9 for comparison). This is likely due to a reduced crystal quality of TMDC layers and organic residues from the transfer process, yet further advances in TMDC growth and transfer techniques are expected to yield improved film quality [49, 50].

Figure 4d summarizes the normalized AlScN (0002) peak intensity on different hexagonal templates. A close correlation between crystal quality and in-plane lattice mismatch is observed: the high crystallinity is achieved on TMDC buffers with similar lattice constants, and decreases with increasing mismatch in both tensile and compressive strain ranges. These results further elucidate the quasi-vdW epitaxy mechanism, and offer an effective strategy for integration of wurtzite ferroelectrics. The family of 2D TMDCs, with varying lattice constants, provides the platform to grow high-quality wurtzite nitrides with varying compositions and on various substrates regardless of lattice symmetry and constant. Importantly, the thickness of TMDC buffers (~0.5 nm for monolayer) is significantly thinner than the conventional metal or nitride buffers, thereby minimizing the vertical dimension and adding flexibility for designing devices (see Table S2 for detailed comparison) [17, 23, 25, 45].

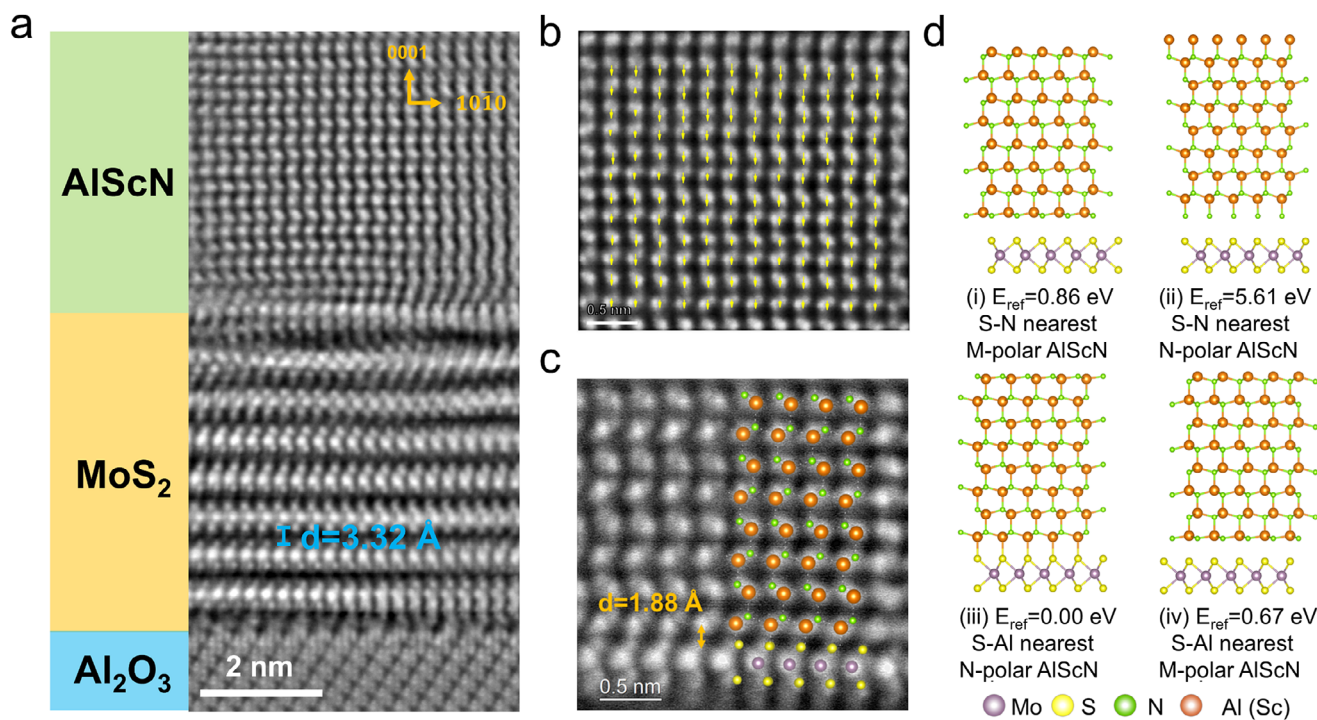


FIGURE 3 | Atomic structure of AlScN/MoS₂ heterostructure. (a) iDPC-STEM image of the AlScN/8-layer MoS₂/Al₂O₃. (b) The mapping of ferroelectric polarization of AlScN. (c) Atomic structure of the AlScN/MoS₂ interface. (d) The possible atomic configurations of the interface and calculated relative formation energy.

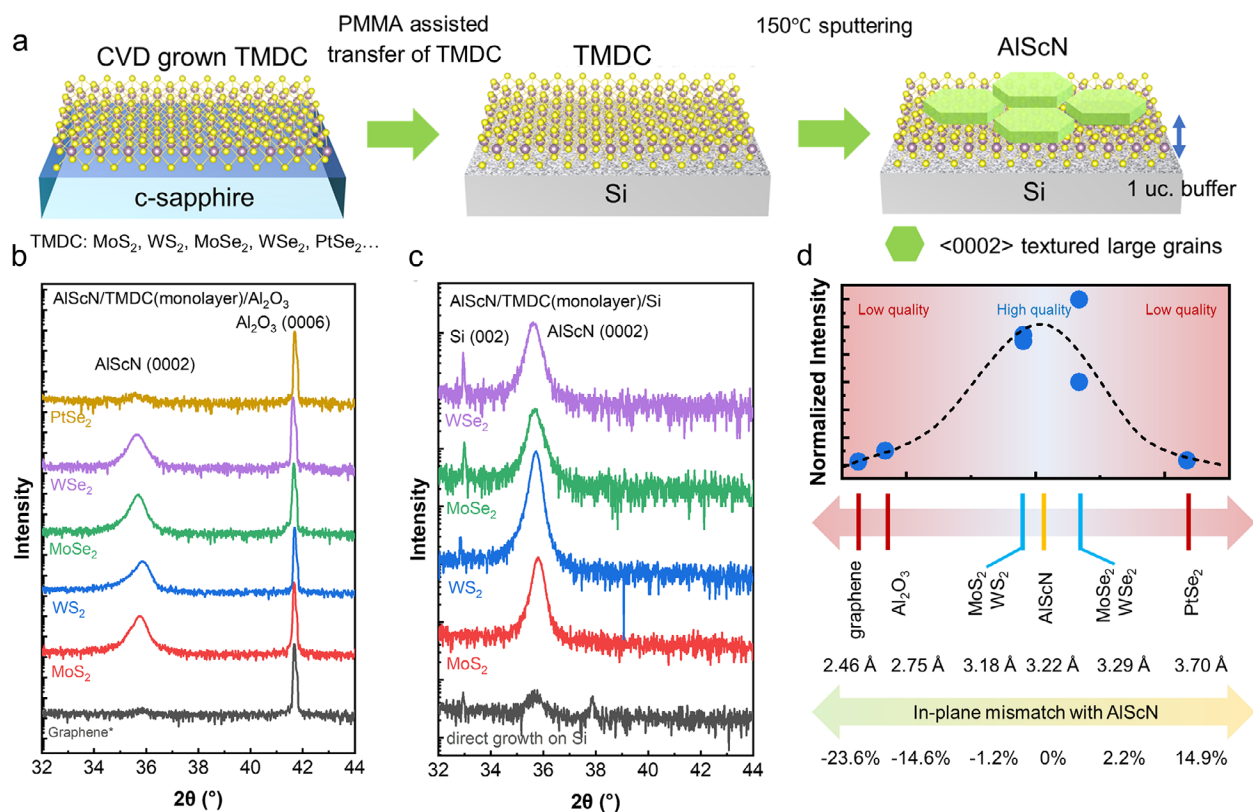


FIGURE 4 | Universality of quasi-vdW epitaxy of AlScN. (a) Schematic diagram of integrating AlScN on different substrates including Si. (b) θ -2 θ diffraction results of 50 nm-thick AlScN grown on Al₂O₃, using various monolayer-thick TMDC materials as buffers. (c) θ -2 θ diffraction results of 50 nm-thick AlScN grown on Si substrates, using different monolayer-thick TMDCs as buffers. (d) Correlation between AlScN (0002) diffraction peak intensity and in-plane lattice constant of different hexagonal templates for AlScN on Al₂O₃.

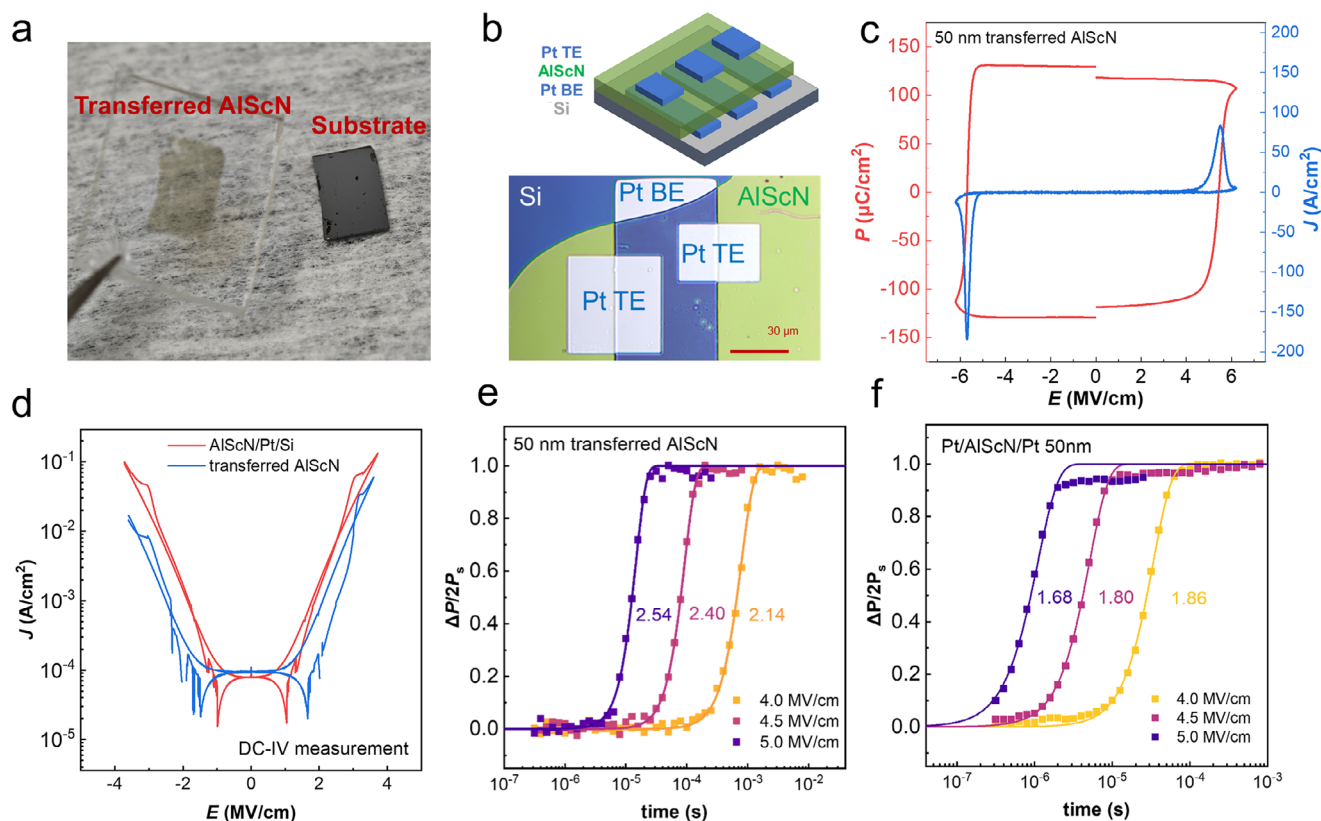


FIGURE 5 | Ferroelectric properties of freestanding AlScN. (a) Photograph of 50 nm AlScN membrane on PDMS, exfoliated from Al₂O₃ substrate. (b) Schematic diagram and optical image of ferroelectric capacitor fabricated by transferring 50 nm AlScN freestanding membrane, with the scale bar of 30 μm. The bottom electrode (BE) was patterned before transfer, while the top electrode (TE) was fabricated by electron beam lithography. (c) Ferroelectric hysteresis loops of the transferred capacitor. The red line corresponds to ferroelectric polarization, while the blue line corresponds to switching current. (d) DC leakage current of the transferred capacitor and the conventional Pt/AlScN/Pt capacitor on Si with the same AlScN thickness. (e,f) Switching kinetics of (e) the transferred capacitor and (f) conventional capacitor characterized by normalized switched polarization versus pulse time. The capacitor was first set to N-polar with positive saturated electric fields, and then measured using square pulses with negative electric field from 4.0 to 5.0 MV/cm.

2.4 | Ferroelectricity of Freestanding AlScN Membranes

The use of TMDC buffers with vdW interactions further enables the fabrication of freestanding AlScN membranes, unlocking the potential of AlScN-based flexible electronics and vdW hetero-integration [34–36]. Using water-assisted exfoliation and PMMA/PDMS transfer strategy (see Methods and Figure S11a), 50 nm freestanding AlScN membranes with a large yield (Figure 5a) and crack-free areas up to hundreds of micrometers scale were successfully obtained (see Figure S11 for further discussion). Moreover, this exfoliation is reliably achieved using multilayer TMDC buffers except for the monolayer TMDC, highlighting the critical role of vdW interaction for exfoliation.

Next, the membranes were transferred onto Si substrates with patterned Pt bottom electrodes, followed by fabricating Pt top electrodes. Figure 5b shows the schematic diagram and optical microscopy image of Pt/AlScN/Pt capacitors developed by this transfer technique (hereafter referred to as transferred capacitors). For comparison, Pt/AlScN/Pt multilayers were also grown directly on Si and fabricated into capacitor devices (hereafter referred to as conventional capacitors, see Methods for fabrication details). Figure 5c presents the electric-field hysteresis loops

of polarization (P - E) and displacement current (J - E) of the transferred capacitors with 50 nm AlScN, measured by using positive-up-negative-down (PUND) method (see full data in Figure S12). The results show a sharp ferroelectric switching with remanent polarization (P_r) around 120 μC/cm², which is higher than that of the conventional capacitors (Figure S13) and consistent with previous reports [6, 51, 52]. Moreover, quasi-static direct current-voltage (DC-IV) measurements reveal that the transferred capacitors exhibit a lower leakage current with ferroelectric switching displacement current at ~3.0 MV/cm (Figure 5d). Note that the different switching fields for DC-IV and PUND (~5.4 MV/cm) are due to the voltage frequency (see Figure S14 for detailed discussion). Moreover, robust ferroelectricity is also demonstrated for freestanding AlScN membranes with thinner thickness (see Figure S15).

To reveal the effect on ferroelectric switching kinetics, we performed transient current measurements [44]. The time-dependent normalized switched polarization for the transferred and conventional capacitors with 50 nm AlScN are shown in Figure 5e,f, respectively. The kinetic processes were fitted by using the Kolmogorov-Avrami-Ishibashi (KAI) model with a variable Avrami exponent n , which represents the effective dimension of domain growth [53, 54]. Notably, the change of n value

reflects variation in the degree of domain wall pinning. For the transferred capacitor, a sharp transition is observed with $n > 2$ across the electric field from 4.0 to 5.0 MV/cm. This is consistent with the previously reported simultaneously nonlinear nucleation growth model, indicating a fast domain wall motion with nonlinear domain nucleation [53, 55]. In contrast, the conventional capacitor shows a gradual switching kinetics with $n < 2$ under the same electric field, suggesting that domain wall motion is pinned by defects or grain boundaries [56].

Our results show that the freestanding AlScN membranes exhibit excellent ferroelectric properties, including high polarization, low leakage current and rapid switching kinetics (see comparison in Table S3 and Figure S16), likely associated with the high crystallinity. These are beneficial for developing ferroelectric memories with high modulation ratio, low power dissipation and fast response [8, 45, 47, 57, 58]. Moreover, the capability of transferring freestanding membranes opens pathways for hetero-integration of high-quality AlScN on various materials, highly desirable for next-generation electronic devices.

3 | Conclusion

In conclusion, we demonstrate a quasi-vdW epitaxy strategy using TMDC monolayers as buffers to achieve low-temperature and high-quality growth of AlScN films. The universality of this strategy is demonstrated by using a wide range of 2D TMDC buffers and on substrates with distinct symmetries. The vdW nature of buffer also enables large-scale exfoliation of freestanding AlScN films, which exhibit excellent ferroelectric properties. We anticipate that this strategy is applicable to a broad family of wurtzite ferroelectric and piezoelectric materials such as AlBN, AlYN, and ScGaN [3]. Moreover, the expanded substrate choice, largely reduced buffer thickness and inherent transferability unlock a new avenue for integrating wurtzite ferroelectrics in next-generation electronic devices, including piezoelectric MEMS filters and actuators, ferroelectric memories, vdW logic devices and nonlinear optics [8, 9, 36, 59, 60].

4 | Experimental Section

4.1 | Sample Fabrication

MoS₂ and other TMDC buffers were grown by CVD on c-plane sapphire (Al₂O₃), and transferred onto Si substrates (provided by SixCarbon Technology Shenzhen). Al_{0.74}Sc_{0.26}N films were deposited by dual-target radio frequency (RF) reactive magnetron sputtering in AJA system using 2-inch-diameter Al and Al_{0.57}Sc_{0.43} targets. The RF power of Al and AlSc targets were 120 and 180 W, respectively. The background pressure was kept below 1×10^{-8} Torr to minimize oxygen incorporation. Prior to deposition, the substrates were degassed in high vacuum at 300°C for 40 min to remove adsorbed oxygen and organic residue. Flow of 3.3 sccm of argon (Ar) and 6.6 sccm of nitrogen (N₂) was introduced, and the total pressure was maintained at 2 mTorr. The growth temperature for AlScN on TMDC buffers was 150°C. For growth of Pt buffers on Si, 60-nm-thick Pt layers were grown after deposition of 5 nm AlN (RF power of 180 W, 2 mTorr), under

atmosphere of 2 mTorr Ar, direct-current (DC) power of 120 W and growth temperature of 400°C. For growth of Pt buffers on Al₂O₃, 60-nm-thick Pt layers were grown under 2 mTorr Ar, DC power of 30 W and growth temperature of 450°C. For growth of AlScN on Pt/Si and Pt/Al₂O₃, the deposition temperature was 350°C, with other conditions identical to the AlScN on TMDC buffers. For fabrication of Pt top electrodes on AlScN, we used room-temperature DC sputtering and patterned by standard lift-off lithography.

4.2 | Transfer Process

A layer of PMMA (950 A4) was spin-coated (2500 rpm and 60 s) onto the AlScN surface to serve as a support layer. The PMMA/AlScN/substrate was baked at 180°C for 3 min on a hot-plate. After baking, the edges of the sample were scratched with knives to allow solution penetration. Then a PDMS intermediate stamp was attached on top of the PMMA and the sample was soaked in deionized water at room temperature for 24 h to weaken the vdW interactions between the TMDC and the substrate. After soaking, a tweezer was used to peel off PDMS/PMMA/AlScN from the original substrate. Following that, a homemade transfer stage was used to transfer the stack onto pre-patterned Pt/Si electrodes. Finally, the PMMA was removed by acetone, and post-annealed at 200°C under high vacuum ($\sim 10^{-4}$ Pa) to improve the electrical contacts. The top electrodes for transferred AlScN were defined by standard electron beam lithography using PMMA as a photoresist.

4.3 | Characterizations

The phase, crystallinity and thickness of thin films were characterized using XRD and XRR, by high-resolution X-ray diffractometer (Empyrean PANalytical) with Cu K_{α1}. Raman spectra were performed on Witec Alpha 300S confocal Raman microscope using 532 nm laser with 5 mW power focused on 50× objective at ambient atmosphere. TEM specimens were prepared using a dual-beam focused ion beam (FIB, Helios 5 UC, and ThermoFisher Scientific). After preparation, the specimens were bonded on commercial Cu grids and thinned down by Ga ions with accelerating voltage and beam current of 30 kV and 2.5 nA. The final thickness for TEM specimens was about 30–40 nm. TEM annular dark field images were obtained through high-angle annular dark-field (HAADF) with acceptance angles of 39–200 mrad. The atomic-scale iDPC-STEM images were investigated by a double spherical-aberration corrected scanning transmission electron microscope (STEM, Spectra 300, and ThermoFisher Scientific). The crystal structures were simulated by VESTA.

4.4 | DFT Calculations

The DFT calculations were performed using the projector augmented plane-wave (PAW) basis set, which was implemented within Vienna ab-initio simulation package (VASP) [61]. The plane-waves were truncated at 500 eV, while the exchange correlations of electrons were described using generalized gradient

approximations (GGA) based on the functional proposed by Perdew, Burke, and Ernzerhof (PBE) [62]. The semiempirical DFT-D3 method parameterizing the van der Waals correction is used in the calculations [63]. The vacuum layer of 20 Å was introduced in the z-direction, deliberately chosen to be nonperiodic in order to effectively eliminate any interactions between adjacent images. A dipole correction was implemented, and a stringent energy convergence criterion of 1×10^{-6} eV was employed for solving the self-consistent Kohn–Sham equations. To ensure accurate sampling of the Brillouin zone, resolutions better than 0.02 Å⁻¹ were used following the Monkhorst–Pack scheme [64]. All the structures in this study, including meticulously designed partially fixed heterostructures, were relaxed until achieving a Hellman–Feynman convergence criterion smaller than 0.01 eV Å⁻¹.

4.5 | Electrical Measurements

Macroscopic ferroelectric measurements were performed by Keithley 4200A-SCS parameter analyzer, with two 4225PMU modules to apply pulsed voltage with minimum duration of 20 ns. The wake-up process of 100 cycles was performed on the ferroelectric capacitors before measurements. The frequency used in hysteresis and PUND tests were 10 kHz. SMU modules were used to measure the leakage current of thin films. Transient current measurements for switching kinetics were conducted by square waveforms, see previous work for details [44]. The integration and fittings were performed using Origin.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that supports the findings of this study are available in the supplementary material of this article.

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