Effects of adatom mobility and Ehrlich– Schwoebel barrier on heteroepitaxial growth of scandium nitride (ScN) thin films

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ABSTRACT

Scandium nitride (ScN) is an emerging rock salt indirect bandgap semiconductor and has attracted significant interest in recent years for thermoelectric energy conversion, as a substrate for defect-free GaN growth, as a semiconducting component in single-crystalline metal/ semiconductor superlattices for thermionic energy conversion, as well as for $Al_{1-x}Sc_xN$ -based bulk and surface acoustic devices for 5G technologies. Most ScN film growth traditionally utilizes physical vapor deposition techniques such as magnetron sputtering and molecular beam epitaxy, which results in stoichiometric films but with varying crystal quality, orientations, microstructures, and physical properties. As epitaxial single-crystalline ScN films with smooth surfaces are essential for device applications, it is important to understand the ScN growth modes and parameters that impact and control their microstructure. In this Letter, we demonstrate that large adatom mobility is essential to overcome the Ehrlich–Schwoebel (E–S) and grain boundary migration barriers and achieve defect (voids, dislocations, stacking faults, etc.)-free single-crystalline ScN films. Using the substrate temperature to tune adatom mobility, we show that nominally single-crystalline ScN films are achieved when the homologous temperature is higher than ~0.3. For homologous temperatures ranging from 0.23 to 0.30, ScN films are found to exhibit significant structural voids in between pyramidal growth regions with multiple in-plane orientations resulting from additional lateral growth off the facets of the pyramids and broken epitaxy after ~80 nm of growth. The in-depth discussion of the growth modes of ScN presented here explains its varying electrical and optical properties and will help achieve high-quality ScN for device applications.

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Transition metal mono-nitrides (TMNs) are researched extensively in recent years for corrosion resistant coating and refractory electronic and plasmonic applications due to their high mechanical hardness and large melting temperatures. Compared to the traditional TMNs such as TiN, ZrN, and CrN, scandium nitride (ScN) is relatively less explored and has emerged as an attractive semiconducting TMN for thermoelectric energy conversion,^{1–3} as a substrate for defect-free GaN growth for light-emitting diodes (LEDs),^{4,5} and as a semiconducting component in ZrN/ScN and TiN/(Al,Sc)N metal/semiconductor superlattice-based thermionic energy conversion devices.^{6–9} ScN is mechanically hard (~24 GPa), is corrosion and oxidation resistant, exhibits a large melting temperature of 2600 °C, and is structurally, morphologically, and chemically stable at ambient conditions.^{10–14} Solid-solution alloys of ScN with other III–V semiconductors such as AlN and GaN have attracted significant attention due to their tunable bandgaps, dielectric permittivity, and electronic properties for high-power applications.^{15–17} Wurtzite-Al_xSc_{1–x}N has also emerged as an attractive piezoelectric material for bulk and surface acoustic resonators in high frequency power electronics and 5G technologies.^{18–20} Mn-doped ScN is predicted to exhibit dilute magnetic semiconducting properties and has the potential for spintronic applications.^{21–23}

Unlike other well-known III-nitride semiconductors, ScN crystallizes in the rock salt structure with octahedral bonding coordination and exhibits an indirect (Γ -X) bandgap of 0.9 eV and a direct (Γ - Γ)

gap of 2.2 eV.^{24–26} The as-deposited ScN films exhibit an *n*-type carrier concentration of $\sim 10^{20} - 10^{21} \text{ cm}^{-3}$ that arises primarily due to the presence of unwanted oxygen.^{27–29} Molecular beam epitaxy (MBE) and sputter-deposited ScN thin films exhibit mobility ranging from 60 to 120 cm²/V s at room temperature with acoustic phonon scattering, impurity scattering, and dislocation scattering dominating at higher temperatures.^{30–33} With a large growth rate of 2–4 μ m/h, ScN films deposited with hybrid vapor phase epitaxy (HVPE) have exhibited the highest reported mobility of ~284 cm²/Vs at room temperature thus far.³⁴ Due to the large mobility and suitable carrier concentrations, both MBE and sputter-deposited ScN films exhibit a high Seebeck coefficient (\sim -150 to -180 μ V/K) and a large thermoelectric power factor of $\sim 2.5 \times 10^{-3}$ W/m K² at 500 K–600 K, which are comparable to many well-established thermoelectric materials such as Bi2Te3 and PbTe.^{32,33,35,36} The large electron concentration in ScN has been reduced with Mg (hole)-doping, and *p*-type ScN thin films have been demonstrated with the maximum hole concentration of 2×10^{20} cm⁻³ and hole mobility of $\sim 20 \text{ cm}^2/\text{V} \text{ s}$ at room temperature.³⁷ Photoemission experiments have also demonstrated a rigid-band electronic structure of ScN for both *n*-type (unwanted oxygen) and *p*-type (intentional Mg) doping, which leads to its large electron and holeconcentrations, large Seebeck coefficients, and thermoelectric power factor across the *n*-type to *p*-type carrier transition regime.³

Most ScN thin films are usually deposited with physical vapor deposition (PVD) techniques such as MBE and magnetron sputtering on various substrates such as Si, MgO, GaN, and Al₂O₃, which results in nearly stoichiometric films but with varying crystalline quality, crystal orientations, microstructures, surface morphology, and roughness.^{28,39-41} Previous research on the impact of deposition pressure on sputter deposited ScN on (001) MgO showed that the crystalline quality and surface morphology of films degrade with an increase in deposition pressure from 2 mTorr to 20 mTorr, leading to a reduction in mobility and thermoelectric power factor.²⁹ Similarly, for MBE deposited ScN on Si substrates, crystalline quality and surface roughness were found to depend strongly on the Sc flux.⁴² However, a detailed understanding of the effects of growth temperature and adatom mobility on the growth mechanism of ScN is still missing. Along with the surface diffusion barrier, when an adatom in the terrace approaches the step edge, it has to overcome the Ehrlich-Schwoebel (E-S) barrier⁴³ that results from changes in the coordination number and is responsible for the uphill growth with the rough surface morphology as found in several other nitride thin films previously.44-47 Since epitaxial defect-free ScN thin films with sub-nm surface roughness are essential for ScN-based Schottky and pn-junction diodes and are used for thermoelectric, optoelectronic, and thermionic energy conversion devices, it is necessary to understand the growth modes and parameters that result in defect-free single-crystalline ScN films. The analysis is further necessitated due to the fact that as-deposited ScN usually contains oxygen as an impurity that acts as a heavy n-type dopant and impacts its electrical properties. However, not much understating exists on the mechanism of oxygen incorporation and the relationship between the oxygen concentration and distribution with the microstructure. Therefore, employing a combination of high-resolution scanning transmission electron microscopy (HRSTEM) and transmission Kikuchi diffraction (TKD), in this Letter, we examine the various growth modes of ScN and demonstrate that high-quality defect-free ScN films are achieved when the adatom mobility overcomes the E-S

and grain boundary migration barriers during deposition. The homologous temperature [ratio between the growth temperature (T) and melting temperature (T_m)] is found to play a critical role in the eventual microstructure, crystal quality, surface morphology, and oxygen concentration and distribution in the film.

Three ScN thin films with substrate temperatures of 600 °C, 700 °C, and 800 °C are deposited with DC-magnetron sputtering inside an ultra-high vacuum chamber with a base pressure of 2×10^{-9} Torr on (001) MgO substrates. The details about the growth process are presented in the supplementary material section. The high resolution symmetric 2θ - ω x-ray diffractogram shows (see Fig. S3 in the supplementary material) that all three ScN films grow with 002 orientations on (001) MgO substrates. From the 002 ScN peak positions, a lattice constant of 4.50 Å is extracted for all three films, which is consistent with previous literature reports.^{32,48} The full-width-athalf-maximum (FWHM) of the rocking curve (ω -scan) is found to be 1.26°, 0.74°, and 0.50° for the films deposited at 600 °C, 700 °C, and 800 °C, respectively. Since the FWHM of the rocking curve is an indication of the crystalline quality of films, the result shows that films deposited at higher temperature exhibit superior crystalline quality.

The microstructure of the ScN film deposited at 600 °C was imaged by cross-sectional HRSTEM (see Fig. 1). Electron diffraction patterns (EDPs) and low-magnification TEM images show that the film grows with the cubic (001)[001] ScN || (001)[001] MgO epitaxial relationship for the first 80–100 nm of the film thickness (region-I)



FIG. 1. (a) Low-magnification cross-sectional STEM image of the ScN film deposited on the MgO substrate at 600 °C showing three different regions. The void lines inclined at $\sim\pm$ 63° to the ScN/MgO interface run along {102} planes as seen from the EDP pattern given in the inset. (b) Enlarged image of the marked portion in (a) where the voids are clearly visible. (c) Voids with positive and negative angles meat and appear as boundaries of triangular pyramidal grains. (d) Void lines originating at ScN/MgO interfaces and running along {102} planes are shown. HRTEM-EDS elemental maps of Sc (e), N (f), and O (g) are presented that show the voids are depleted of Sc and N atoms, but rich in oxygen atoms along the boundaries.

beyond which defects appear. Subsequent to the appearance of defects, two regions are identified: region-II, where though the film still maintains the same epitaxial relationship with the substrate but contains defect lines that run $\sim \pm 63^{\circ}$ with respect to the substrate (MgO) surface, and region-III, where the epitaxy breaks completely and appears dark in HRTEM images as it is out of the zone axis. The signatures of the broken epitaxy can be seen as weak spots between the square patterns in the electron diffraction pattern (EDP) (presented in Fig. S2). Some areas exhibit region-II spanning through the sample and terminating at the surface rendering flat square shaped structures, while in most of the film, region-III terminates at the surface with a pyramidlike structure on the surface as seen in the plan-view scanning electron microscopy (SEM) image [see Fig. 4(a)]. STEM-EDS (energy dispersive x-ray spectroscopy) analysis shows that the defect lines appearing at $\sim \pm 63^{\circ}$ with respect to the substrate surface are structural voids separating regions of pyramidal growth and are depleted of Sc and N, but rich in oxygen atoms at the edges [see STEM-EDS elemental mapping along such voids in Figs. 1(e)-1(g)]. Closer inspection reveals that the voids originate at the ScN/MgO interface [see Fig. 1(d)] and run along {102} planes throughout the film as evident from the EDP shown in Fig. 1(a). Several voids are seen parallel to each other, and the voids forming $+63^{\circ}$ and -63° to the substrate/film interface eventually meet each other, thus forming pyramids [see Fig. 1(c)].

To understand the orientations of the grains in region-III, TKD analysis was performed on the TEM cross-sectional lamellae. The pole figure shows the brightest spots at the center and at the four edges corresponding to {100} planes in the epitaxial regions aligning with the X, Y, and Z directions of the TKD map [see Fig. 2(a)], as expected. Apart from the main {100} planes, four more types of orientations can be identified from the figure corresponding to region-III that are tilted by 56°-60° with respect to the substrate [clearly seen in Fig. 2(b)] and indicate how far the 001 plane is from the Z axis. Inverse pole figure (IPF) maps [see Fig. 2(c)] show the real-space distribution of the four orientations located between {101} and {111} planes in the Y direction. The clusters of grains span a range of orientations as shown in the pole figure; however, the majority are within 5° of the 122 plane for the Z axis of the IPF Z map. Orientation angle distribution over 25 grains shows preferential orientations of the grains, suggesting that they grow off the facets of the pyramids, effectively creating a second growth front until they meet the respective laterally adjacent grain. Though the layer-by-layer growth is hindered in region-II, the epitaxy and orientation remain the same as region-I, evidenced by the inverse pole figure maps [Fig. 2(c)].

The origin of the microstructure and determination of the growth conditions for defect-free ScN is discussed based on the systematic zone model for physical vapor deposition that relates deposition parameters such as substrate temperature, pressure, and flux to the microstructure.^{49–52} According to this model, thin films deposited with the physical vapor deposition theed different zones based on the homologous temperature (T/T_m).⁵³ In zone-I, where the homologous temperature is less than 0.1, the adatoms have negligible mobility and stick to the film surface wherever they arrive. Due to the low mobility and kinetic energy, the adatoms cannot overcome the surface diffusion



FIG. 2. (a) Transmission Kikuchi diffraction (TKD) pole figure of the ScN film deposited on the MgO substrate at 600 °C that shows the main spots corresponding to the (001) growth of ScN epilayers on the (001) MgO substrate. There are 12 lower intensity spots arising due to tilted grains in region III, which are more clearly shown in (b). (b) Four sets of additional spots (marked with different circles) that are tilted at 56°–60° with respect to the interface arising from the grains of region-III, where the broken epitaxy is vivid after negating the (001) spot. (c) Inverse pole figure maps corresponding to each of the four sets of peaks showing four different types of orientations of the broken epitaxy region that are oriented between {101} and {111}.



FIG. 3. Systematic zone model for physical vapor deposited films that relates the microstructures to the adatom mobility. Low adatom mobility and inability to overcome the surface diffusion barrier in zone-I lead to porous films with voids. The Ehrlich–Schwoebel (E–S) barrier in zone-T leads to island growth and epitaxy breaking with multiple orientations. When the adatoms have sufficient mobility (energy) to overcome the E–S and grain boundary migration barrier, columnar growth is achieved in zone-II. (b) One-dimensional schematic of the diffusion barrier and E–S barrier encountered by an adatom at the terrace edge. (c) Voids are formed along the grain boundaries (gray region) due to the shadowing of the arriving adatoms by the already formed triangular grains.

barrier, which results in a high density of island structures found in the initial stage of the growth. The island growth results in columnar or pyramidal structures with voids due to the self-shadowing effect [Fig. 3(a)]. As the substrate temperature is increased, the adatoms gain sufficient energy to overcome the surface diffusion barrier. However, as the adatoms move along the surface layer, they encounter the Ehrlich–Schwoebel (E–S) barrier that results from the necessary changes in the coordination number for downward movement of adatoms at the step edges [see Fig. 3(b)].^{54,55} The E–S barrier prevents interlayer diffusion and results in vertical growth of grains, resulting in triangular (pyramidal) shapes. With the adatom mobility not sufficient for the grain boundary migration, fresh grains start to develop on the side facets of non-coalesced triangular grains with different orientations. Since these grains grow on the rough surfaces of non-coalesced grains and on the higher index planes, their growth rates vary significantly from the initial film growth rate. Such microstructures formed when the adatom mobility (energy) is insufficient to overcome the E–S barrier and the activation energy required for grain boundary migration are classified as zone-T [see Fig. 3(a)] with the homologous temperature ranging from 0.10 to 0.30. A further increase in growth temperature (with a homologous temperature greater than 0.30) results in enough adatom mobility for the interlayer diffusion and grain boundary migration that results in wide columnar growth of films in zone-II.

For ScN, the film deposited at 600 °C corresponds to a homologous temperature of 0.23 that falls under the zone-T, and that is why the microstructure observed in STEM images in Fig. 1 contains three different regions: epitaxy intact region (I), epitaxial yet voided region (II), and broken epitaxy region (III), as predicted by the model. The triangular grains formed due to the E–S barrier and the secondary grains growing from the valleys of the former grains, blocking the admission of adatoms in between the grains as depicted in Fig. 3(c), resulting in the formation of voids. The voids formed along the grain boundaries are oriented $\sim \pm 63^{\circ}$ to the substrate most probably as a result of stress relaxation along the low energy plane.⁵⁶

Therefore, to achieve defect-free epitaxial ScN, the substrate temperature was increased to 700 °C (corresponding to a homologous temperature of 0.27) and 800 °C (corresponding to a homologous temperature of 0.31 and the maximum substrate temperature achievable with the deposition system). Plan-view SEM images (see Fig. 4) show that although the film deposited at 700 °C falls under zone-T, the crystal quality improves significantly with fewer triangular grains on the surface compared to the film deposited at 600 °C. However, for the film deposited at 800 °C, a clear transition from zone-T to zone-II was observed with the surface being nearly free from triangular grains. Low- and high magnification (S)/TEM images and EDS maps (see Fig. 5) verify that the film deposited at 800 °C is epitaxial and nominally single-crystalline throughout its whole thickness, with only a single vertical defect line being visible along the entire field of view [Fig. 5(a)]. Sharp and abrupt ScN/MgO interfaces are found in the HRSTEM image [see Fig. 5(b)], while due to the \sim 6%–7% latticemismatch between the film and the substrate, some misfit dislocations are also visible. It is interesting to note that even though dislocations are present at the interface, the film does not exhibit voids as verified with the elemental Sc and O maps that show a homogeneous and



FIG. 4. Plan-view SEM images of the ScN thin films deposited at (a) 600 °C, (b) 700 °C, and (c) 800 °C. The density of triangular pyramid structures seen on the surface of the film deposited at 600 °C reduces as the deposition temperature is increased, eventually resulting in a smooth film at 800 °C. The magnified images are given in the inset with the scale bar corresponding to 100 nm.



FIG. 5. (a) Low magnification TEM image of the ScN film deposited on the MgO substrate at 800 °C. Electron diffraction pattern in the inset confirms the epitaxial growth and the absence of grains with different orientations in contrast to the films deposited at lower temperatures. (b) High angle annular dark field-scanning transmission electron microscopy image of the ScN/MgO interface showing cubic epitaxy and the presence of misfit dislocations due to lattice mismatch between ScN and MgO. STEM-EDS elemental mapping of (c) Sc and (d) O shows homogeneous elemental distribution.

continuous distribution across the film [see Figs. 5(c) and 5(d)]. AFM measurements (see the supplementary material) showed that the rms roughness of the films deposited at 700 °C and 800 °C was 4.3 nm and 1 nm, respectively, representing the improvement in surface smoothness with an increase in the substrate temperature.

Apart from the growth modes of ScN, the STEM-EDS mapping is employed for quantitative estimation of the oxygen concentration to address the oxygen incorporation mechanism in ScN. For the film deposited at 600 °C, the oxygen concentrations were found to be 8 at. %, 12 at. %, and 18 at. % in regions I, II, and III, respectively [see Fig. 1(a)]. In the earlier reports on ScN growth, the oxygen incorporation was identified to occur during the growth and not post deposition.^{28,41} However, in this study, as the Sc source metal contains \sim 0.5 at. % of oxygen as impurity and given the base pressure was 2×10^{-9} Torr prior to deposition, it is reasonable to hypothesize that the large oxygen concentration in the ScN film deposited at 600 °C results from post deposition oxygen diffusion from the atmosphere along the edges of the voids as seen in Fig. 1(g). For the film deposited at 800 °C, the oxygen concentration was found to be much lower at 2 at. % and is distributed uniformly throughout the film with no accumulation of excessive oxygen along the voids. Therefore, the analysis establishes the previously assumed hypothesis that the amount of oxygen in the ScN film depends on its crystalline quality and a low oxygen content can be achieved with lower defect density in the films.⁴⁷ However, a detailed study on the oxygen incorporation mechanism inside ScN remains to be performed. Room-temperature electrical and optical properties such as carrier density, mobility, and conductivity as well as optical transition and bandgap are further measured and presented in the supplementary material, which exhibit correlation with the microstructure of ScN.

In conclusion, using the substrate temperature during deposition to tune the adatom mobility, we demonstrate that epitaxial defect-free ScN thin films are achieved only when the adatoms possess sufficient mobility to overcome the Ehrlich–Schwoebel (E–S) and grain boundary migration barriers. For adatom energy (mobility) less than that of the barrier heights, the films contain structural voids that are depleted of Sc and N atoms due to the self-shadowing effect and the epitaxy of the film breaks after \sim 80–100 nm of growth. Oxygen impurities are found in a much larger concentration, clustered around the voids and defects. The homologous temperature of the growth, therefore, plays a critical role in determining the morphology, crystalline quality and orientation, surface smoothness, and incorporation of oxygen in the film as an impurity. The present work will help understand different growth modes of ScN in physical vapor deposited films and will help develop epitaxial defect-free single-crystalline ScN for electronic, opto-electronic, and thermoelectric device applications.

See the supplementary material for information related to growth, characterization methods, high resolution x-ray diffraction and AFM analysis, electrical measurements, and optical spectroscopy analysis.

AUTHORS' CONTRIBUTIONS

D.R. and B.B. contributed equally to this work.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

REFERENCES

- S. Kerdsongpanya, N. Van Nong, N. Pryds, A. Žukauskaite, J. Jensen, J. Birch, J. Lu, L. Hultman, G. Wingqvist, and P. Eklund, Appl. Phys. Lett. 99, 232113 (2011).
- ²A. Shakouri, Annu. Rev. Mater. Res. 41, 399 (2011).
- ³S. Kerdsongpanya, B. Alling, and P. Eklund, J. Appl. Phys. **114**, 73512 (2013).
 ⁴M. A. Moram, Y. Zhang, M. J. Kappers, Z. H. Barber, and C. J. Humphreys, Appl. Phys. Lett. **91**, 152101 (2007).
- ⁵Proceedings of the First Symposium on III-V Nitride Materials and Processes, edited by T. D. Moustakas, J. P. Dismukes, and S. J. Pearton (The Electrochemical Society Inc., Pennington, NJ, 1996), Vol. 96, p. 197.
- ⁶V. Rawat, Y. K. Koh, D. G. Cahill, and T. D. Sands, J. Appl. Phys. **105**, 024909 (2009).
- ⁷B. Saha, Y. R. Koh, J. Comparan, S. Sadasivam, J. L. Schroeder, M. Garbrecht, A. Mohammed, J. Birch, T. Fisher, A. Shakouri, and T. D. Sands, Phys. Rev. B 93, 045311 (2016).
- ⁸M. Garbrecht, L. Hultman, M. H. Fawey, T. D. Sands, and B. Saha, J. Mater. Sci. 53, 4001 (2018).
- ⁹M. Garbrecht, I. McCarroll, L. Yang, V. Bhatia, B. Biswas, D. Rao, J. M. Cairney, and B. Saha, J. Mater. Sci. 55, 1592 (2020).
- ¹⁰K. A. Gschneidner, Jr., *Rare Earth Alloys* (Van Nostrand, Princeton, New Jersey, 1961), OCLC: 1705458.
- ¹¹G. V. Samsonov, M. D. Lyutaya, and V. S. Netspor, Zh. Prikl. Khim. **36**, 2108 (1963), [Russ. J. Appl. Chem. **36**, 2043 (1963)], OSTI: 4134244.
- ¹²D. Gall, I. Petrov, N. Hellgren, L. Hultman, J. E. Sundgren, and J. E. Greene, J. Appl. Phys. 84, 6034 (1998).
- ¹³J. P. Dismukes, W. M. Yim, and V. S. Ban, J. Cryst. Growth 13-14, 365 (1972).
- ¹⁴B. Biswas and B. Saha, Phys. Rev. Mater. **3**, 020301 (2019).
- ¹⁵S. Zhang, D. Holec, W. Y. Fu, C. J. Humphreys, and M. A. Moram, J. Appl. Phys. **114**, 133510 (2013).
- ¹⁶H. C. L. Tsui, L. E. Goff, S. K. Rhode, S. Pereira, H. E. Beere, I. Farrer, C. A. Nicoll, D. A. Ritchie, and M. A. Moram, Appl. Phys. Lett. **106**, 132103 (2015).
- ¹⁷ H. C. L. Tsui, L. E. Goff, R. G. Palgrave, H. E. Beere, I. Farrer, D. A. Ritchie, and M. A. Moram, J. Phys. D 49, 265110 (2016).
- ¹⁸W. Wang, P. M. Mayrhofer, X. He, M. Gillinger, Z. Ye, X. Wang, A. Bittner, U. Schmid, and J. K. Luo, Appl. Phys. Lett. **105**, 133502 (2014).
- ¹⁹Y. Lu, Development and Characterization of Piezoelectric AlScN-Based Alloys for Electroacoustic Applications (University of Freiburg, Freiburg im Breisgau, 2019).
- ²⁰M. Akiyama, T. Kamohara, K. Kano, A. Teshigahara, Y. Takeuchi, and N. Kawahara, Adv. Mater. 21, 593 (2009).
- ²¹H. A. Al-Brithen, H. Yang, and A. R. Smith, J. Appl. Phys. **96**, 3787 (2004).
- ²²B. Saha, G. Naik, V. P. Drachev, A. Boltasseva, E. E. Marinero, and T. D. Sands, J. Appl. Phys. **114**, 063519 (2013).
- ²³A. Herwadkar and W. R. L. Lambrecht, Phys. Rev. B 72, 235207 (2005).
- ²⁴B. Saha, J. Acharya, T. D. Sands, and U. V. Waghmare, J. Appl. Phys. 107, 033715 (2010).
- ²⁵X. Bai and M. E. Kordesch, Appl. Surf. Sci. 175-176, 499 (2001).
- ²⁶M. S. Haseman, B. A. Noesges, S. Shields, J. S. Cetnar, A. N. Reed, H. A. Al-Atabi, J. H. Edgar, and L. J. Brillson, APL Mater. 8, 081103 (2020).

- ²⁷H. Al-Atabi, Q. Zheng, J. S. Cetnar, D. Look, D. G. Cahill, and J. H. Edgar, Appl. Phys. Lett. **116**, 132103 (2020).
- ²⁸M. A. Moram, Z. H. Barber, and C. J. Humphreys, Thin Solid Films **516**, 8569 (2008).
- ²⁹P. V. Burmistrova, D. Zakharov, T. Favaloro, A. Mohammed, E. Stach, A. Shakouri, and T. Sands, J. Mater. Res. **30**, 626 (2015).
- ³⁰T. Ohgaki, K. Watanabe, Y. Adachi, I. Sakaguchi, S. Hishita, N. Ohashi, and H. Haneda, J. Appl. Phys. **114**, 093704 (2013).
- ³¹J. S. Cetnar, A. N. Reed, S. C. Badescu, S. Vangala, H. A. Smith, and D. C. Look, Appl. Phys. Lett. **113**, 192104 (2018).
- ³²P. V. Burmistrova, J. Maassen, T. Favaloro, B. Saha, S. Salamat, Y. Rui Koh, M. S. Lundstrom, A. Shakouri, and T. D. Sands, J. Appl. Phys. **113**, 153704 (2013).
- ³³D. Rao, B. Biswas, E. Flores, A. Chatterjee, M. Garbrecht, Y. R. Koh, V. Bhatia, A. I. K. Pillai, P. E. Hopkins, M. Martin-Gonzalez, and B. Saha, Appl. Phys. Lett. **116**, 152103 (2020).
- ³⁴Y. Oshima, E. G. Villora, and K. Shimamura, J. Appl. Phys. 115, 153508 (2014).
- ³⁵J. Wei, L. Yang, Z. Ma, P. Song, M. Zhang, J. Ma, F. Yang, and X. Wang, J. Mater. Sci. 55, 12642 (2020).
- ³⁶P. Eklund, S. Kerdsongpanya, and B. Alling, J. Mater. Chem. C 4, 3905 (2016).
- ³⁷B. Saha, M. Garbrecht, J. A. Perez-Taborda, M. H. Fawey, Y. R. Koh, A. Shakouri, M. Martin-Gonzalez, L. Hultman, and T. D. Sands, Appl. Phys. Lett. 110, 252104 (2017).
- ³⁸S. Nayak, M. Baral, M. Gupta, J. Singh, M. Garbrecht, T. Ganguli, S. M. Shivaprasad, and B. Saha, Phys. Rev. B 99, 161117 (2019).
- ³⁹A. R. Smith, H. A. H. Al-Brithen, D. C. Ingram, and D. Gall, J. Appl. Phys. **90**, 1809 (2001).
- ⁴⁰J. Casamento, J. Wright, R. Chaudhuri, H. Xing, and D. Jena, Appl. Phys. Lett. 115, 172101 (2019).
- ⁴¹A. L. Febvrier, N. Tureson, N. Stilkerich, G. Greczynski, and P. Eklund, J. Phys. D 52, 035302 (2019).
- ⁴²M. A. Moram, C. J. Humphreys, S. V. Novikov, A. J. Kent, C. T. Foxon, and C. Nörenberg, J. Cryst. Growth **310**, 2746 (2008).
- ⁴³S. J. Liu, H. Huang, and C. H. Woo, Appl. Phys. Lett. **80**, 3295 (2002).
- ⁴⁴H. Turski, F. Krzyżewski, A. Feduniewicz-Zmuda, P. Wolny, M. Siekacz, G. Muziol, C. Cheze, K. Nowakowski-Szukudlarek, H. (Grace) Xing, D. Jena, M. Załuska-Kotur, and C. Skierbiszewski, Appl. Surf. Sci. 484, 771 (2019).
- ⁴⁵K. C. Maurya, V. M. Shalaev, A. Boltasseva, and B. Saha, Opt. Mater. Express 10, 2679 (2020).
- ⁴⁶H. N. Shah, Int. J. Automot. Mech. Eng. 14, 3872 (2017).
- ⁴⁷J. More-Chevalier, S. Cichoň, L. Horák, J. Bulíř, P. Hubík, Z. Gedeonová, L. Fekete, M. Poupon, and J. Lančok, Appl. Surf. Sci. 515, 145968 (2020).
- ⁴⁸D. Gall, I. Petrov, L. D. Madsen, J.-E. Sundgren, and J. E. Greene, J. Vac. Sci. Technol., A 16, 2411 (1998).
- ⁴⁹J. A. Thornton, J. Vac. Sci. Technol., A 4, 3059 (1986).
- 50 I. Petrov, P. B. Barna, L. Hultman, and J. E. Greene, J. Vac. Sci. Technol., A 21, S117 (2003).
- ⁵¹P. G. S. Mahieu, D. Depla, and R. De Gryse, Thin Solid Films 515, 1229 (2006).
 ⁵²A. Anders, Thin Solid Films 518, 4087 (2010).
- ⁵³B. A. Movchan and A. V. Demchishin, Fiz. Met. Metalloved. 28, 653 (1969), OSTI: 4181669.
- 54G. Ehrlich and F. G. Hudda, J. Chem. Phys. 44, 1039 (1966).
- ⁵⁵R. L. Schwoebel and E. J. Shipsey, J. Appl. Phys. 37, 3682 (1966).
- ⁵⁶M. Garbrecht, L. Hultman, M. H. Fawey, T. D. Sands, and B. Saha, Phys. Rev. Mater. 1, 033402 (2017).