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ABSTRACT

Two $In_xAl_{1-x}N$ layers were grown simultaneously on different substrates [sapphire (0001) and the Ga-polar GaN template], but under the same reactor conditions, they were employed to investigate the mechanism of strain-driven compositional evolution. The resulting layers on different substrates exhibit different polarities and the layer grown on sapphire is N-polar. Moreover, for the two substrates, the difference in the degree of relaxation of the grown layers was almost 100%, leading to a large In-molar fraction difference of 0.32. Incorporation of In in $In_xAl_{1-x}N$ layers was found to be significantly influenced by the strain imposed by the under-layers. With the evolutionary process of In-incorporation during subsequent layer growth along [0001], the direction of growth was investigated in detail by Auger electron spectroscopy. It is discovered that the $In_{0.60}Al_{0.40}N$ layer grown directly on sapphire consists of two different regions with different molar fractions: the transition region and the uniform region. According to the detailed cross-sectional transmission electron microscopy, the transition region is formed near the hetero-interface due to the partial strain release caused by the generation of misfit-dislocations. The magnitude of residual strain in the uniform region decides the In-molar fraction. $In_xAl_{1-x}N$ layers were analyzed by structural and optical characterization techniques. Our present work also shows that a multi-characterization approach to study $In_xAl_{1-x}N$ is a prerequisite for their applications as a buffer layer.

INTRODUCTION

As a tunable bandgap (E_g) from the infrared (InN, $E_g \sim 0.7$ eV) to the deep ultraviolet (AlN, $E_g \sim 6.2$ eV), ternary $In_xAl_{1-x}N$ has great potential for optoelectronic and electronic devices.[1–5] This further extends to strain engineering by tuning of lattice-parameters from the largest InN ($a = 3.54$ Å, $c = 5.76$ Å) to the smallest AlN ($a = 3.11$ Å, $c = 4.98$ Å) either to improve material quality or electronic properties. For example, $In_{0.17}Al_{0.83}N$ lattice matched to GaN leads to a reduction in the number of defects and has been used as a barrier layer since the last two decades for applications in high electron mobility transistors (HEMTs), light-emitting diodes, laser diodes, solar cells, and highly reflective distributed Bragg reflectors.[6–10] In 2011, Kuzmík and Georgakilas[11] have proposed a potential usage of the In-rich $In_xAl_{1-x}N$ thick layer as a buffer layer for strained InN-channel HEMTs, which could be promising for high frequency applications.

Growing the high quality $In_xAl_{1-x}N$ layer (either $x \sim 0.17$ or $x > 0.17$) by means of the organometallic chemical vapor deposition

(OMCVD) technique has always been a challenge due to the substantial dissimilarities between physical and chemical parameters of binary nitrides, AlN and InN.[12,13] The growth of the $In_xAl_{1-x}N$ alloy over the entire composition range was pioneered by Starosta[14] using reactive multi-target sputtering on microscope glass slides in 1981. Guo *et al.*[15] has grown $In_xAl_{1-x}N$ in the whole composition range directly on (0001) sapphire by reactive radio-frequency magnetron sputtering in 2003. This is probably owing to the fact that sputtering is a physical rather than a chemical deposition technique. Consequently, thermodynamic equilibrium conditions are not required for the growth of the $In_xAl_{1-x}N$ layer. Subsequently, a number of other techniques have also been reported to grow $In_xAl_{1-x}N$ on the conventional GaN buffer layer that include molecular beam epitaxy (MBE)[16–18] and horizontal organometallic chemical vapor deposition (OMCVD).[19–21] GaN as a buffer layer is an obvious choice for $In_xAl_{1-x}N$ growth because it is used already for high power and high frequency devices. However, parasitic gallium auto-incorporation into the $In_xAl_{1-x}N$ layer due to the memory effect of GaN base layer growth is another reported issue.[22]

To our knowledge, magnetron sputtering has been the only reported technique for the $In_xAl_{1-x}N$ growth on (0001) sapphire and their optical properties have also been studied.[15,23–25] Nevertheless, a detailed structural investigation is still missing. In the present work, we compared two $In_xAl_{1-x}N$ layers grown simultaneously under the same growth conditions by OMCVD but on different substrates: (0001) c-plane sapphire and the GaN template. We systematically investigated the epitaxial growth conduction by means of a high-resolution X-ray diffraction (HR-XRD) method, high-resolution transmission electron microscopy (HRTEM), and scanning transmission electron microscopy (STEM) with atomic resolution. HR-XRD was used to characterize the composition and strain in the $In_xAl_{1-x}N$ layers. In-depth In-composition was analyzed by Auger electron spectroscopy (AES). Our study of $In_xAl_{1-x}N$ on sapphire provides considerable insight into crystal layer growth, which can be translated onto other rhombohedral substrates as well.

EXPERIMENT

Samples in this study were grown by low pressure OMCVD, using an AIXTRON $3 \times 2''$ flip-top (FT) close coupled showerhead (CCS) reactor. Trimethylindium (TMIn), trimethylaluminum (TMAl), and ammonia (NH_3) were utilized as In, Al, and N precursors, respectively. N_2 was used as a carrier gas. The schematic cross section of two samples is shown in Figs. 1(a) and 1(b). The only difference between samples A and B lies in the under-layers; however, the growth conditions for $In_xAl_{1-x}N$ layers were the same. For sample A, $In_xAl_{1-x}N$ was grown directly on sapphire. On the other hand, $In_xAl_{1-x}N$ was grown on the GaN template ($\sim 2\,\mu$m GaN grown on sapphire at 1100 °C, then the reactor was baked and cleaned prior to the $In_xAl_{1-x}N$ growth) in sample B. For both samples, $In_xAl_{1-x}N$ layers were grown in one run. Prior to the $In_xAl_{1-x}N$ growth initiation, substrates were exposed to NH_3 flow for 300 s at 1011 °C. Following nitridation, the growth temperature was ramped down to 730 °C in preparation for $In_xAl_{1-x}N$ layer growth. Prior to the $In_xAl_{1-x}N$ layer growth for 8440 s, an AlN layer growth was carried out for 30 s. The other reactor conditions used to grow $In_xAl_{1-x}N$ layers were a 7×10^3 Pa pressure, a 5087 V/III ratio, and a 10 slm total gas flow rate. Growth temperature and layer thickness were monitored *in situ* using LayTec EpiCurve®TT optical (632.7 nm) reflectance.

HR-XRD measurements were performed using the Bruker D8 DISCOVER diffractometer equipped with a rotating Cu anode and operating at 12 kW. Diffractions 0004 and $11\bar{2}4$ of $In_xAl_{1-x}N$ were measured to determine molar fractions, lattice parameters, and strain states. A linear dependence of relaxed lattice parameters (a_r, c_r) and elastic coefficients (c_{13}, c_{33}) of $In_xAl_{1-x}N$ on the composition x was employed in the calculation. In the evaluation, the values $a_{GaN} = 0.318907$ nm, $c_{GaN} = 0.51855$ nm (PDF 00-50-0792), $a_{AlN} = 0.311117$ nm, $c_{AlN} = 0.498017$ nm (PDF 03-65-0832), and $a_{InN} = 0.35378$ nm, $c_{InN} = 0.57033$ nm (PDF 00-50-1239) were used as initial lattice parameters of GaN, AlN, and InN crystals, respectively. The elastic properties of AlN and InN were described by the stiffness constants $c_{13}^{AlN} = 103$ GPa, $c_{33}^{AlN} = 373$ GPa, $c_{13}^{InN} = 92$ GPa, and $c_{33}^{InN} = 224$ GPa, respectively, calculated by density functional-theory.[26] The density of threading dislocations (TDs) in the layers was estimated from the widths (FWHM, β) of the 0002 and $10\bar{1}1$ rocking curves (RCs) measured in symmetric and skew geometry, respectively. ϕ-scans were measured for the $10\bar{1}1$ diffraction of $In_xAl_{1-x}N$ and $10\bar{1}4$ and $11\bar{2}3$ diffractions of the (0001) sapphire substrate in the range of $0° - 360°$ to assess the orientation relation between $In_xAl_{1-x}N$ lattice and sapphire. The surface morphology of samples was evaluated by using NTEGRA Prima atomic force microscope (AFM) and FEI Quanta 250 FEG scanning electron microscope (SEM). Cross-sectional SEM was also performed to verify the as-grown layers thickness. Elemental depth profile was obtained by a Jeol JAMP 9510-F Auger microprobe (using 10 keV electron excitation with 10 nA beam current, 55° tilt, and 1 keV Ar^+ ions employed for sputtering) taking advantage of its excellent surface sensitivity and identification of light elements. Due to the lack of standard samples, matrix relative sensitivity factor was not known, and correspondingly, the y-axis was calibrated in the Auger-peak-to-peak height (APPH) fraction using equal relative sensitivity factors.[27] HRTEM was carried out in an FEI CUBED TITAN microscope equipped with a monochromator and a spherical aberration (Cs) corrected objective lens. It allows chemical analysis using electron energy loss spectroscopy (EELS) and energy dispersive X-ray spectroscopy (EDX). The negative Cs mode, which permits

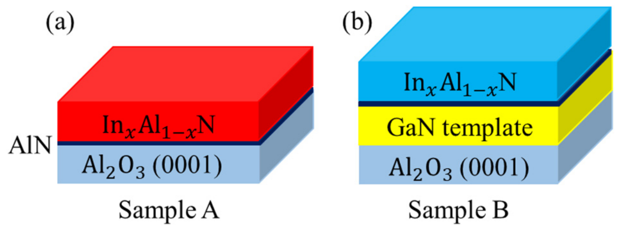

FIG. 1. (a) Schematic layer structure illustration for sample A. The deposition was initiated with nitridation, followed by a 1–2 nm-thick HT-AlN nucleation layer and a 334 nm-thick $In_xAl_{1-x}N$ layer. (b) Schematic layer structure illustration for sample B. The layer structure consists of a $\sim 2\,\mu$m-thick GaN template grown on the Al_2O_3 (0001) and $In_xAl_{1-x}N$ layer.

one to image also light elements, has been used to determine the polarity of the layers in the case of growth on sapphire, which cannot be used as a reference for convergent beam diffraction as it is centro-symmetric. The analysis of the layers crystallographic quality was carried out in the weak beam mode using a JEOL 2010 microscope. Polarized point Raman spectra were measured in $z(xx)\bar{z}$ geometry to observe long-wavelength optical phonon mode behavior by using Renishaw InVia™ Raman spectrometer in the confocal mode with 488 nm excitation laser wavelength and an ∼270 nm spot size on the sample. Optical properties were investigated by photoluminescence (PL) and optical absorbance (OA) employing the OLYMPUS BX51 microscope equipped with He-Cd laser (325 nm) and Shimadzu UV-VIS-NIR Spectrophotometer SolidSpec-3700, respectively.

RESULTS AND DISCUSSION

The In-incorporation in the $In_xAl_{1-x}N$ layer was approximately two times higher when grown on sapphire (In-molar fraction 0.60, sample A) compared with the GaN template (In-molar fraction 0.28, sample B). Figure 2 shows a typical 2θ/ω XRD scan of sample A observed in a broad range from 30.5° to 36.5°. The full width at half maximum (FWHM) of the $In_{0.60}Al_{0.40}N$ 0002 peak observed at 33.07° in the 2θ/ω scan is 0.20°. No diffraction peaks are observed at the positions expected for pure InN 0002 at 31.33° and for pure AlN 0002 at 36.14°, which indicates that the obtained 334 nm thick $In_{0.60}Al_{0.40}N$ layer has a single-phase structure and desired growth orientation. The diffraction pattern indicates a heteroepitaxial relationship between $(0001)_{In_{0.60}Al_{0.40}N}$ and $(0001)_{Al_2O_3}$. The rocking curve (ω-scan) of $In_{0.60}Al_{0.40}N$ 0002 and $10\bar{1}1$ diffractions has a FWHM value of 0.20° and 0.77°, respectively. The corresponding screw and edge dislocation densities calculated according to Refs. 27 and 28 are 2×10^9 cm^{-2} and 1×10^{11} cm^{-2}, respectively. This indicates that the $In_{0.60}Al_{0.40}N$ layer reported here has better quality than other published MBE grown layers of the same In-molar fraction with FWHM of 0.51°[18] and 0.67°[29] measured by the ω-scan of 0002 diffraction.

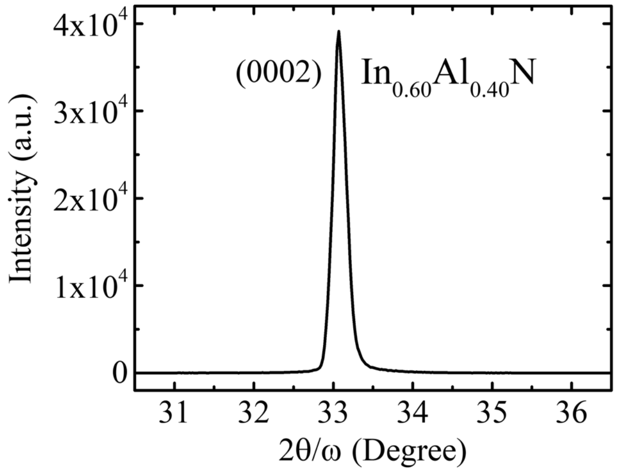

FIG. 2. Representative XRD 2θ/ω scan of $In_{0.60}Al_{0.40}N$ grown on (0001) sapphire.

In the case of sample A, the degree of in-plane alignment of $In_{0.60}Al_{0.40}N$ relative to the (0001) Al_2O_3 substrate was determined by evaluation of ϕ-scans as given in Fig. 3. Three ϕ-scans, $10\bar{1}4$ and $11\bar{2}3$ of the (0001) Al_2O_3 substrate and $10\bar{1}1$ of $In_{0.60}Al_{0.40}N$ layer, were measured in the whole 360° range of the azimuthal

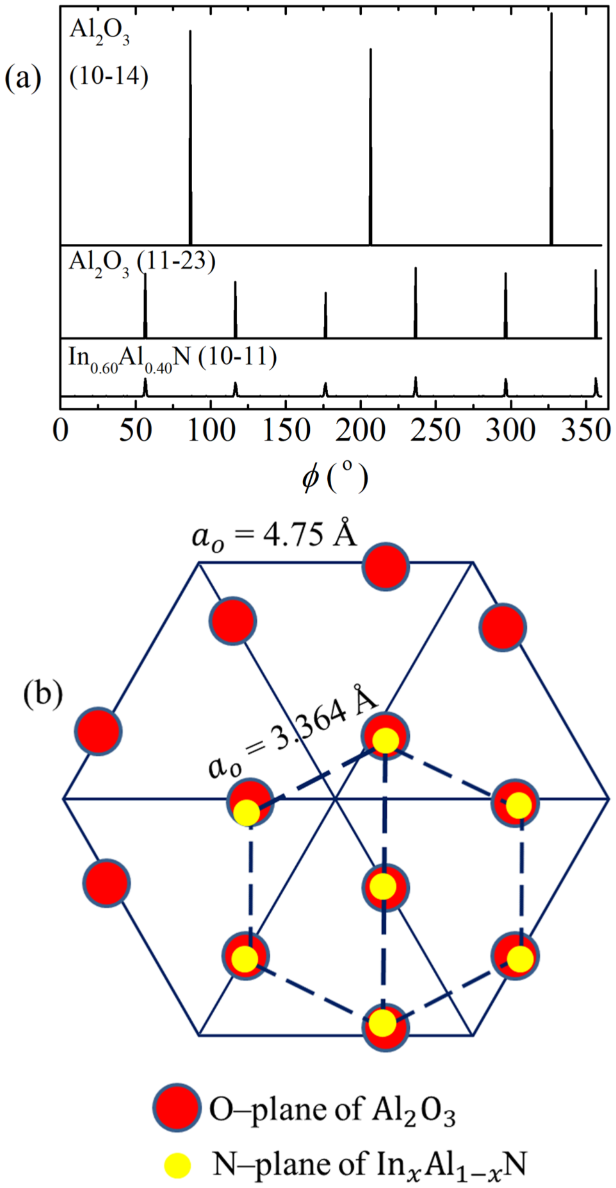

FIG. 3. (a) XRD ϕ-scans of $10\bar{1}4$ and $11\bar{2}3$ diffraction of the (0001) Al_2O_3 substrate and $10\bar{1}1$ diffraction of the grown $In_{0.60}Al_{0.40}N$ layer. (b) Schematic drawing of epitaxial matching relation between Al_2O_3 and $In_xAl_{1-x}N$ at the interface.

angle. Note the threefold symmetry of the $10\bar{1}4$ scan that reflects the rhombohedral symmetry of the Al_2O_3 lattice. It is seen that the positions of the $10\bar{1}1$ maxima perfectly coincide with the maxima of the $11\bar{2}3$ scan of the substrate indicating that the in-plane direction $[10\bar{1}0]$ of $In_{0.60}Al_{0.40}N$ is parallel with the direction $[11\bar{2}0]$ of Al_2O_3. This means that the $In_{0.60}Al_{0.40}N$ lattice is 30° rotated with respect to Al_2O_3, which is the same rotation as observed for GaN on Al_2O_3,[30] and hence, the same applies to sample B. One can summarize the in-plane heteroepitaxial relationship between $In_xAl_{1-x}N$ grown on Al_2O_3 as $In_xAl_{1-x}N(0001)[10\bar{1}0]\|Al_2O_3(0001)[11\bar{2}0]$, and we assume that this relationship holds for the whole composition range of $In_xAl_{1-x}N$. Schematic representation of the interface between the (0001) Al_2O_3 substrate and the InAlN layer is shown in Fig. 3(b).

The designed samples A and B exhibit a large strain difference in $In_xAl_{1-x}N$ layers as a consequence of using different underlayers. The precise values of x were calculated from the positions of 0004 and $11\bar{2}4$ diffractions in reciprocal space determined by HR-XRD. Hexagonal c-oriented epitaxial layers are transversely isotropic, i.e., the deformation of the layer is completely described by two non-zero strain components,

$$\varepsilon_{\|}(x) = \frac{a - a_r(x)}{a_r(x)}, \qquad \varepsilon_{\perp}(x) = \frac{c - c_r(x)}{c_r(x)}, \tag{1}$$

where a, c and a_r, c_r are the lattice parameters of strained (and measured) and fully relaxed layer, respectively. These components are connected by the relation

$$\varepsilon_{\perp} = -2\frac{c_{13}(x)}{c_{33}(x)}\varepsilon_{\|}. \tag{2}$$

It has to be pointed out that in addition to the relaxed lattice parameters a_r, c_r, the aspect ratio c_r/a_r and the elastic properties expressed by c_{13}/c_{33} are functions of the composition x as indicated above. Therefore, Eqs. (1) and (2) for unknown values of a_r, c_r, $\varepsilon_{\|}$, and x have to be solved numerically. An efficient and simple iterative procedure for this solution was proposed in Chauhan *et al.*[27] It was also shown that for ternary compounds, the solution is unique for the given input parameters a and c determined experimentally. Using this procedure, the layer composition x for samples A and B was precisely determined taking correctly into account the in-plane strain component $\varepsilon_{\|}$. For the measured and calculated in-plane lattice parameters a and a_r, the degree of relaxation R is standardly evaluated as

$$R = \frac{a - a_{sub}}{a_r - a_{sub}}, \tag{3}$$

where a_{sub} is the in-plane lattice parameter of the substrate.

For sample A, the values $\varepsilon_{\|} \sim 0$ and $R \sim 1$ were obtained, i.e., the layer is completely relaxed. In the case of sample B, however, the strain in the $In_xAl_{1-x}N$ layer is −1.4% and $R \sim 0$, which means that the strain in the $In_xAl_{1-x}N$ layer is compressive and is almost completely preserved. Considering that $In_xAl_{1-x}N$ layers in samples A and B were grown under the same growth conditions, it is reasonable to assume that a large In-molar fraction difference strongly depends on the kind of the substrate used in the experiment and is closely connected with the residual strain in the layer.

Unlike on polar substrates (SiC), the polar orientation is not obvious in the case when the layer is grown on non-polar substrates (Al_2O_3). In this study, the polarity of samples was determined by two techniques: convergent beam electron diffraction (CBED) and negative Cs HRTEM. The high resolution analysis with negative Cs has been especially carried out on sample A to determine the polarity, which cannot be determined by convergent beam electron diffraction, because the layer contains a high density of defects. On the other hand for sample B, the polarity is determined by convergent beam diffraction on the GaN template and InAlN followed the same polarity. Under the growth conditions described in the Experiment section, we obtained $In_xAl_{1-x}N$ layers of both types of polarities. The N-polar $In_xAl_{1-x}N$ layer was obtained for the layer grown on nitridated Al_2O_3 for 300 s at 1011 °C. However, the $In_xAl_{1-x}N$ layer of sample B, grown on the GaN template, was In/Al-polar. A HRTEM image of the N-polar $In_xAl_{1-x}N$ layer of sample A is displayed in Fig. 4; this negative Cs image shows the bond orientation and clearly demonstrates the N-polarity. The observed influence of long and high temperature (HT) nitridation and of the hetero-interface on the $In_xAl_{1-x}N$ layer polarity is consistent with the earlier reported investigations, and detailed explanation can be found elsewhere.[31–33]

The high vapor pressure and following the low growth efficiency of nitrogen (N) could lead to the formation of the $InN - In_2O_3$ alloy, and resulting bandgap discrepancies have been

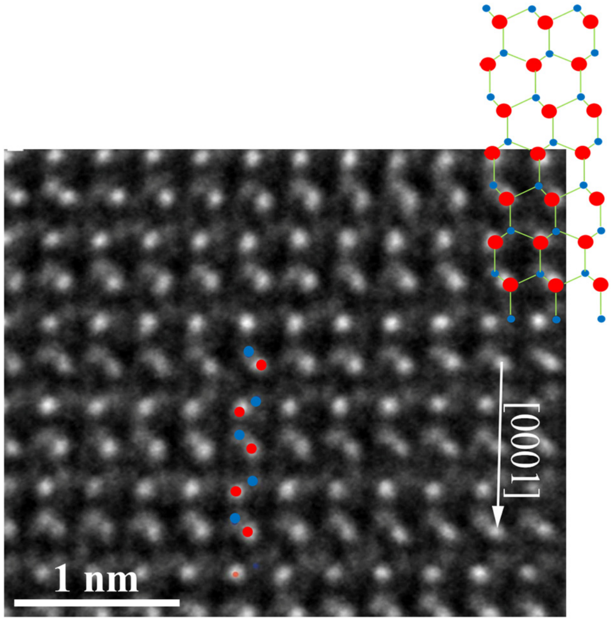

FIG. 4. Negative Cs HRTEM image of the $In_xAl_{1-x}N$ layer of sample A showing aligned columns of N atoms (blue) and In/Al atoms (red). The layer is N-polar; for this image, the Cs was set to −40 μm.

reported for InN. In order to track the In, N, Al, oxygen (O), and carbon (C) profiles of samples A and B along the [0001] direction, AES depth profile was measured and the typical AES profiles are shown in Fig. 5. Due to the mixing of atoms sputtered from a very thin AlN layer with the atoms originating from adjacent layers at the high sputtering rates during AES depth profiling, the 1–2 nm thick AlN layer, as shown in Fig. 1, is not visible in AES profiles of samples. Evidently, the $In_xAl_{1-x}N$ layer of sample A consists of two different regions with different molar fractions: a transition region (light blue region) and a uniform region. In the transition region, In- and Al-molar fractions vary, while they remain nearly the same in the uniform region. For sample A, the Al-molar fraction falls and the complementary In-molar fraction rises rapidly in the initial stage of growth, which corresponds to a 50 nm thick transition region (at the depth ≥275 nm). With the progression of growth, the In-molar fraction enters into the uniform region with a In-molar fraction of 0.60. The transition region occupies a comparatively small part in $In_xAl_{1-x}N$ layers; therefore, the residual strain determined by HR-XRD can be approximately equivalent to the residual strain in the uniform region. The observed decreasing profile of oxygen in the transition region along the growth direction could be attributed to its out-diffusion from sapphire.[34,35] Carbon content close to surface is mainly due to environmental contamination. AES observations indicate that the layer grown on sapphire is homogeneous, considering the measured in-depth distribution of the elements and has an insignificant amount of oxygen impurity.

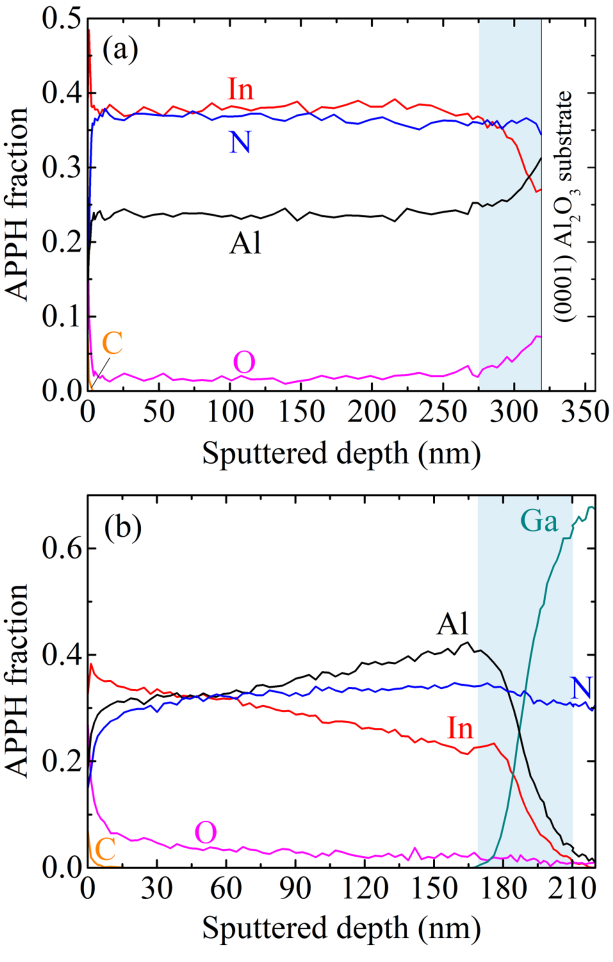

FIG. 5. In, N, Al, O, and C elemental depth profiles in $In_xAl_{1-x}N$ layers along the growth direction [0001] for (a) sample A and (b) sample B. Thickness of $In_xAl_{1-x}N$ layers of samples A and B is approximately 334 nm and 210 nm, respectively. The light blue area is corresponding to the transition region.

In the case of sample B, the $In_xAl_{1-x}N$ layer growth mechanism seems to be additionally complicated due to the existing Ga profile in the transition regions, which could be out-diffused from the underlying GaN layer.[22,36] We did not observe unwanted Ga in the $In_xAl_{1-x}N$ layer of sample A that ruled out growth environment and lingering precursors for being the source of Ga in the transition region of sample B.[37] It is found that In and Al-incorporation in a 40 nm thick transition region is complementary to the diminishing Ga profile in the subsequent layer growth. On the contrary to sample A, no uniform region is observed in sample B. This could be attributed to the 17 nm root-mean-square (RMS) surface roughness of the layer that may influence depth resolution and leads to the increasing interface widening with sputtered depth. With the progression of growth, we observe a gradual and slow increase of In-incorporation. Quantitatively, for $In_xAl_{1-x}N$ layers of samples A and B, the large difference of degree of relaxation close to 100% could lead to a large In-molar fraction difference of 0.32 in the uniform region. The compressively strained layer on the GaN template results with a 0.28 In-molar fraction, which is close to the previous studies on $In_xAl_{1-x}N$[38] and $In_xGa_{1-x}N$[39] under certain growth conditions. These phenomena can be elucidated by the compositional pulling effect (CPE) guided by compressive strain.[40]

The observed different thicknesses or growth rates of $In_xAl_{1-x}N$ layers of samples A (∼0.04 nm/s) and B (∼0.02 nm/s) indicate that not only In-incorporation, but also that the growth rate is influenced by different under-lying substrates. Approximately two times higher growth rate and the In-molar fraction of the $In_xAl_{1-x}N$ layer of sample A compared with sample B indicate that In-incorporation efficiency depends on the growth rate, which is also consistent with the previous reports on $In_xGa_{1-x}N$[41] and $In_xAl_{1-x}N$.[42] Typically, the high defect density in a layer arises from the large residual in-plane strain that is generated by the large lattice mismatch to the used substrate (will be discussed in the TEM section), and consequently, the relaxed $In_xAl_{1-x}N$ layer of sample A could provide a favorable accommodation to the larger size of In adatoms. According to Keller *et al.*,[43] polarity of layers influence In-incorporation as well, whereas in our previous reports,[27,44] we observed that strain relaxation is accompanied by enhanced In-incorporation on In/Al-polar InAl(Ga)N/GaN layers. Therefore, the extent of effectiveness of polarity still needs to be demonstrated. In the following, we will focus exclusively on the experimental analysis of the In-rich $In_xAl_{1-x}N$ layer grown directly on sapphire (sample A), which is the sample of our interest.

Cross-sectional TEM analysis was performed to analyze the growth evolution of $In_xAl_{1-x}N$ on sapphire and subsequently identify dislocations and the possible origin of surface defects. Figure 6 shows a weak beam dark-field image recorded with **g** = 0002, where

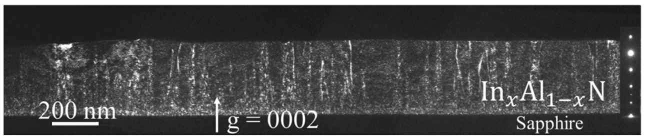

FIG. 6. Weak beam dark-field cross-sectional TEM image of $In_xAl_{1-x}N$ on sapphire: $\mathbf{g} = 0002$.

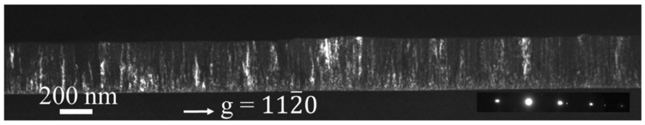

FIG. 8. Weak beam dark-field cross-sectional TEM image of $In_xAl_{1-x}N$ on sapphire: $\mathbf{g} = 11\bar{2}0$.

the interface and the surface of the layer are seen edge-on. In this figure, the following important features can be noticed: First, on the top of the sapphire substrate, a slightly bright contrast is visible in the $In_xAl_{1-x}N$ layer with an extension of around 40 nm, which means that there is a high density of defects. It is worth mentioning here that the 1–2 nm AlN layer could be too thin to have an effect on strain; nevertheless, it promotes strain relaxation in uncovered areas. On the top of this area, the individual threading dislocations with the **c** component are clearly visible and they propagate to the layer surface. Some of them can be seen to induce visible change at the layer surface probably through the formation of pinholes. Finally, the layer surface is not completely flat even at this scale. With an estimate of an average thickness of ~100 nm, there are visible 36 dislocations in this image, which gives a density of around $2.6 \times 10^{10}\ cm^{-2}$.

The picture is more complex in weak beam images recorded using $\mathbf{g} = 10\bar{1}0$ (Fig. 7), where the bright contrast at the $In_xAl_{1-x}N$/Sapphire interface is increased. This is a clear indication that this interface area contains the highest density of defects especially with displacement vectors (Burgers vectors) inside the basal planes. Such defects may be **a** and **a** + **c** as well as stacking faults.[45] Specifically, the stacking faults are not visible when using $\mathbf{g} = 0002$ or $\mathbf{g} = 11\bar{2}0$, which agrees with the lower contrast exhibited in the interfacial area in the corresponding images. Such stacking faults would be easily identified using the high resolution transmission electron microscopy as has been reported earlier.[46,47]

On top of the nucleation layer, it becomes difficult to determine the density of the threading dislocations, which is much higher than in Fig. 6 and majority of dislocations are of **a** type. A close examination of the figure shows that there are many bright bands extended from the top of the interface to the surface. These areas correspond to the walls of **a** threading dislocations, which are so close that cannot be separated in this mode of imaging.[48] The micrograph recorded with $\mathbf{g} = 11\bar{2}0$ is shown in Fig. 8 and also presented the contrast at the interface but with a lower density, which is a strong indication of the presence of stacking faults close to the interface. Bright bands of closely spaced **a** type threading dislocations are also clearly exhibited, and the density of these dislocations is extremely high with lines possible to distinguish towards the layer surface.

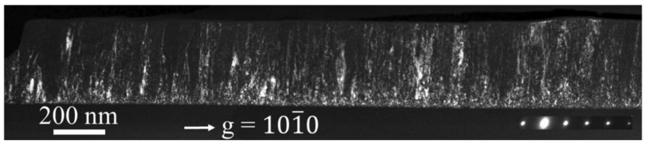

FIG. 7. Weak beam dark-field cross-sectional TEM image of $In_xAl_{1-x}N$ on sapphire: $\mathbf{g} = 10\bar{1}0$.

The relaxation of the misfit strain has been investigated for the growth of the wurtzite GaN buffer layer on sapphire, theoretically[49] as well as experimentally by OMCVD[50] and MBE epitaxy.[51] In the case of conventional buffer GaN on sapphire, the 16% lattice mismatch has been shown to relax rapidly, mainly inside the buffer layer through the formation of multiple types of extended crystallographic defects including basal[50] and prismatic stacking faults[47] but also a strong probability of inversion domain formation.[52,53] Such high defect densities are mainly confined very close to the interface and with a fast decrease of the defect density up to $6\text{–}7 \times 10^8\ cm^{-2}$ at layer thicknesses of around 0.5 μm.[52] In this report, with 22% lattice mismatch on sapphire, the $In_{0.60}Al_{0.40}N$ layer is expected to become relaxed very quickly (the theoretical critical thickness is ~0.76 nm), which should be expected to lead to the localization of most of the strain very close to the interface. However, the density of extended defects inside the epitaxial layer is extremely high, probably in the range of $10^{11}\ cm^{-2}$ as seen above. The detailed investigation of this growth evolution is still underway and will be reported separately.[54]

To investigate the influence of different substrates on surface morphology, $In_xAl_{1-x}N$ layers were analyzed by AFM and SEM measurements. Figure 9 presents the AFM and SEM images of the surface of the $In_xAl_{1-x}N$ layer grown on sapphire. The $In_xAl_{1-x}N$ layer exhibits grain-like surface morphology with flat tops that wrap around dislocations, which is a characteristic of the 2D step-flow (step height $\geq$ 2 MLs) growth mode on top of 3D islands. The observed surface morphology is identical to that of InN grown by MBE,[55,56] being induced by long and high-temperature nitridation and is usually attributed to N-polar layers.[31] The root-mean-square (RMS) surface roughness is ~1 nm over the scan area of $1 \times 1\ \mu m^2$, which is lower than previously reported by Guo *et al.*[15,23] However, RMS roughness of the $In_xAl_{1-x}N$ layer grown on the GaN template is ~17 nm. It is thus proposed that the growth mechanism is changed in the $In_xAl_{1-x}N$ layer grown directly on sapphire because of different surface kinetics.

The quantitative analysis was performed using scanning probe microscopy (SPM) data visualization and analysis tool Gwyddion.[57] From the topography, an average pinhole density of 2.6 (±1.48) × $10^9\ cm^{-2}$ and an average RMS roughness of 5.95 ± 1.4 nm were estimated over a $2.5 \times 2.5\ \mu m^2$ scanned area. Besides low-roughness granular morphology, distinct planar grains with a relatively increased height of 20–24 nm (area) and dislocations decorated at

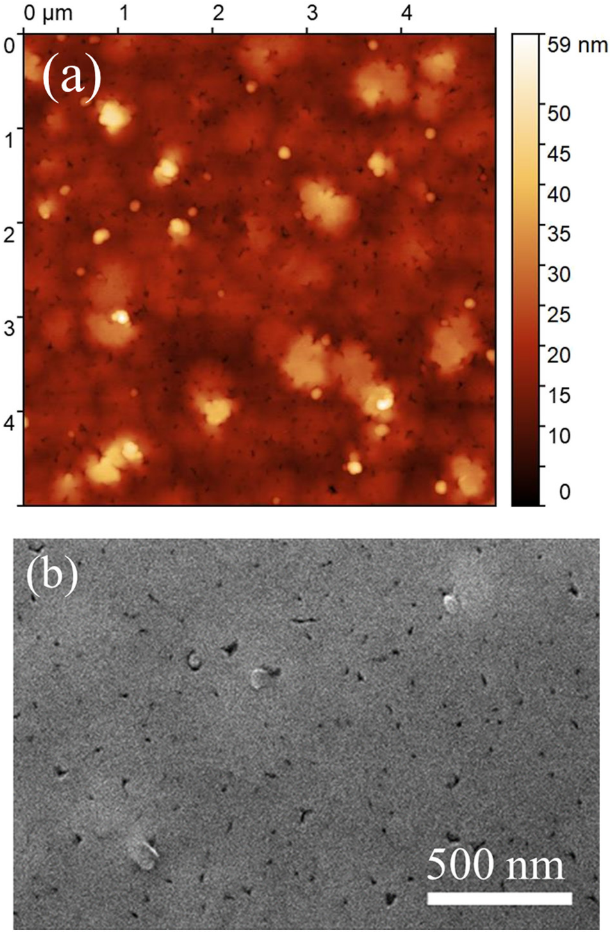

FIG. 9. (a) AFM and (b) SEM images of the surface of the $In_xAl_{1-x}N$ layer on sapphire.

their boundaries are observed. Over a scan area of $12 \times 12\,\mu m^2$, these grains account for ∼25% of the total area with the grain mean height ∼20 ± 1 nm for 44% of grain population and 20–30 nm of the rest. The surface diameter of the grains varies between 200 and 500 nm for 80%. These characteristics show some similarities with the schematic growth of the GaN epilayer, which has been described previously to understand the presence of edge-type threading dislocations at the coalesced grain boundaries of nano-islands with small twist angles.[58] From the TEM and X-ray rocking curve analysis, this is confirmed that most of the dislocations are of edge-type. Finally, since the surface can be described as granular, the numerically calculated height-height correlation function (HHCF) can be employed for the quantitative estimation of surface roughness and lateral correlation length (Fig. 10). The HHCF can be described using a Gaussian distribution defined as

$$\mathrm{HHCF}(X) = 2\sigma^2[1 - \exp(-X^2/\lambda^2)], \tag{4}$$

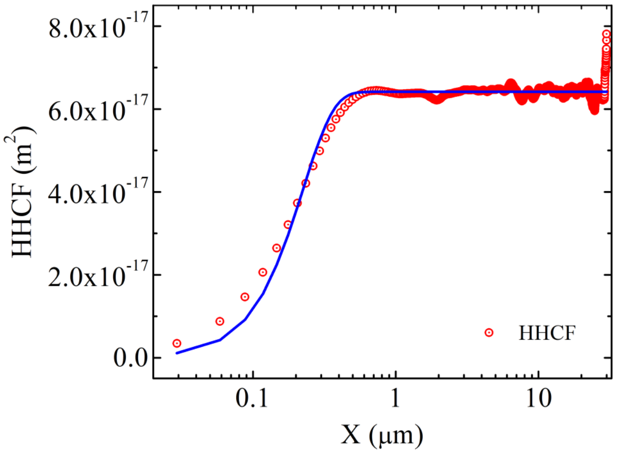

FIG. 10. HHCF variation with one of the spatial coordinates X for the $In_xAl_{1-x}N$ layer on sapphire. Solid blue line shows the fitting of the overall experimental data (red open circles).

where σ and λ are RMS roughness and lateral correlation length, respectively. This analysis was applied over an area of $30 \times 30\,\mu m^2$, and from the fitting of the correlation function, the parameters $\sigma = 5.66$ nm and $\lambda = 223.60$ nm were extracted. The roughness extracted is of similar magnitude as the roughness is calculated in a conventional way. The correlation length would roughly correspond here to the average grain size.

To validate the inherent optical properties of the as-grown $In_xAl_{1-x}N$ layer on sapphire, the absorption edge (AE) energy of the sample was determined by extrapolating the almost linear portion of the optical-absorbance spectrum, as presented in Fig. 11. A non-parabolic conduction band could be the probable reason behind the non-linear absorbance plot.[18] The estimated value of 1.68 eV for the intrinsic bandgap energy of the $In_{0.60}Al_{0.40}N$ layer is consistent with the bandgap of layers fabricated by MBE[18] and OMCVD.[59] However, the $In_{0.60}Al_{0.40}N$ layer is lower than the layers (∼2.8 eV) grown by reactive RF magnetron sputtering[15,23] and RF-MBE,[29] which are close to the theoretical bandgap of ∼2.88 eV estimated by using Vegard's law. It is worth mentioning that the layer bandgap remains unaffected by the presence of negligible amount of oxygen impurity as demonstrated in Fig. 5(a). The OA spectrum is complemented by a sharp, symmetric PL emission peak of $In_{0.60}Al_{0.40}N$ located at 1.79 eV (FWHM = 0.20 eV) as shown in Fig. 11. An insignificant amount of Stokes shift of ∼0.11 eV is observed that is, however, lower than the width of the PL peak. We assume that this disagreement could be because of random local fluctuations of a locally alloyed layer. The deviation from Vegard's law could be defined by the bowing parameter b = 4.53 (for PL) and b = 5.15 (for OA) after using a 0.7 eV InN bandgap and a 6.14 eV AlN bandgap.

To complete the structural characterization of the $In_xAl_{1-x}N$ layer grown directly on sapphire, which is a less studied system and likewise experimental Raman scattering data, we performed Raman spectroscopy. In the Raman spectrum, single-mode behavior of

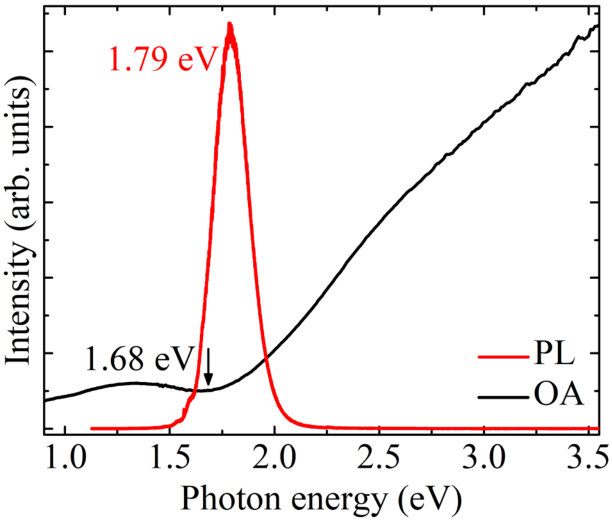

FIG. 11. Room-temperature optical spectra of the $In_{0.60}Al_{0.40}N$ layer measured by different techniques: optical-absorbance (OA) and photoluminescence (PL).

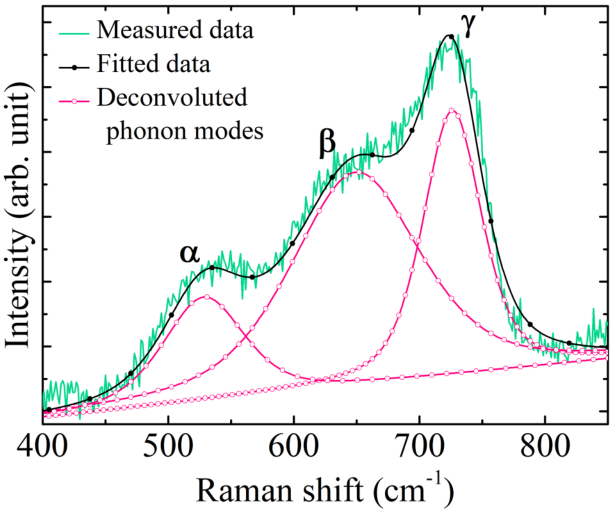

FIG. 12. Room-temperature Raman spectra of the $In_{0.60}Al_{0.40}N$ layer obtained in $z(xx)\bar{z}$ geometry. α, β, and γ consecutively denote E_2(high), InN-like A_1(LO), and AlN-like A_1(LO) phonon modes.

E_2(high) phonon (529 cm^{-1}) and dual-mode behavior of A_1(LO) phonon, InN-like (650 cm^{-1}) and AlN-like (726 cm^{-1}), are observed for sample A as displayed in Fig. 12. The observed two-mode behavior of the A_1(LO) phonon in the In-rich $In_{0.60}Al_{0.40}N$ layer agrees well with the works of Kang *et al.*[20,60] and Tangi *et al.*[61] After using the linear relation between the phonon frequency and composition as reported by Kang *et al.*[20,60] as a reference, we conclude that the E_2(high) (FWHM = 58 cm^{-1}) is red-shifted by 8 cm^{-1}. On the other hand, InN-like (FWHM = 121 cm^{-1}) and AlN-like A_1(LO) (FWHM = 63 cm^{-1}) phonon frequencies are blue-shifted by 13 and 4 cm^{-1}, respectively. According to the theoretical and experimental investigations of Davydov *et al.*[62] and Bergman *et al.*,[63] the phonon energies depend on the Al-molar fraction in $Al_xGa_{1-x}N$ and shift towards higher frequencies with increasing Al-molar fraction. This further leads to the phonon line broadening due to elastic scattering of phonons by fluctuations in randomly disordered composition. Phonon frequency shift could also be attributed to the phonon frequency dependence on free carrier concentration and electric field.[64,65] Oliva *et al.*[66] observed a bowing in the E_2(high) frequency relationship with the In-molar fraction in $In_xGa_{1-x}N$ and showed that the frequency of the A_1(LO) phonon mode is strongly influenced by the in-depth composition of gradients/strain. In the present work, we conclude that the measured red-shifted E_2(high) frequency of the In-rich $In_{0.60}Al_{0.40}N$ layer exhibits a single-mode behavior and suffers from the bowing effect. Furthermore, we conclude that the measured A_1(LO) frequency of the In-rich $In_{0.60}Al_{0.40}N$ layer exhibits two-mode behavior; InN-like A_1(LO) frequency is blue-shifted when compared with AlN-like A_1(LO) due to the In-rich layer and consequently, more broadened due to elastic scattering of phonons by the random disordered compositional fluctuations. The extent of compositional fluctuations in this sample is still under study and will be reported elsewhere.[54] Further research on the role of the $In_xAl_{1-x}N$ buffer layer is needed to investigate electrical properties.

CONCLUSION

We provide experimental evidence of correlation between In-incorporation and strain in the N-polar $In_xAl_{1-x}N$ layer that can promote the growth of a thick In-rich $In_xAl_{1-x}N$ layer applicable to a variety of promising applications or, more specifically, as a buffer in the InN channel transistor for THz applications. The different substrates, sapphire and the GaN template, strongly affect the strain-relaxation mechanism of $In_xAl_{1-x}N$ layers resulting in an increase of the In-molar fraction from 0.28 for the GaN template to 0.60 for sapphire and growth rate from ∼0.02 nm/s to ∼0.04 nm/s, respectively. The $In_xAl_{1-x}N$ layer on sapphire was fully relaxed. On the contrary, the $In_xAl_{1-x}N$ layer grown on the GaN template was completely compressively strained. For the $In_{0.60}Al_{0.40}N$ layer, we observed an optical bandgap variation from 1.68 to 1.79 eV, as measured by OA and PL, respectively. The In-rich $In_{0.60}Al_{0.40}N$ layer also demonstrated single-mode behavior of E_2(high) phonon and dual-mode behavior of A_1(LO) phonon, InN-like and AlN-like. Along with the above-mentioned characterizations that confirmed a good quality of a 334 nm thick N-polar $In_xAl_{1-x}N$ polycrystalline layer grown on sapphire, we proposed a novel approach to grow In-rich $In_xAl_{1-x}N$.

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