Strain-stress study of Al$_x$Ga$_{1-x}$N/AlN heterostructures on c-plane sapphire and related optical properties

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This work presents a systematic study of stress and strain of Al$_x$Ga$_{1-x}$N/AlN with composition ranging from GaN to AlN, grown on a c-plane sapphire by metal-organic chemical vapor deposition, using synchrotron radiation high-resolution X-ray diffraction and reciprocal space mapping. The c-plane of the Al$_x$Ga$_{1-x}$N epitaxial layers exhibits compressive strain, while the a-plane exhibits tensile strain. The biaxial stress and strain are found to increase with increasing Al composition, although the lattice mismatch between the Al$_x$Ga$_{1-x}$N and the buffer layer AlN gets smaller. A reduction in the lateral coherence lengths and an increase in the edge and screw dislocations are seen as the Al$_x$Ga$_{1-x}$N composition is varied from GaN to AlN, exhibiting a clear dependence of the crystal properties of Al$_x$Ga$_{1-x}$N on the Al content. The bandgap of the epitaxial layers is slightly lower than predicted value due to a larger tensile strain effect on the a-axis compared to the compressive strain on the c-axis. Raman characteristics of the Al$_x$Ga$_{1-x}$N samples exhibit a shift in the phonon peaks with the Al composition. The effect of strain on the optical phonon energies of the epitaxial layers is also discussed.

III-Nitride alloys have attracted considerable attention in a wide range of applications of optical, optoelectronic, high-power, and high-frequency devices such as light emitting diodes (LEDs), laser diodes, and high electron mobility transistors (HEMTs)$^{1,6}$. For instance, the hexagonal Al$_x$Ga$_{1-x}$N is one of the most promising candidates for ultraviolet (UV)-LED applications, especially because of its wide bandgap (E$_g$) range from 3.42 eV (for GaN) to 6.2 eV (for AlN) at room temperature$^7$. Al$_x$Ga$_{1-x}$N is also an optimum intermediate layer for InGaN-based LEDs and InAlN transistors$^{2,8-10}$. Al$_x$Ga$_{1-x}$N/GaN HEMTs pave the way for achieving high power radio frequency (RF) devices due to high electron mobility, large critical breakdown field, high sheet charge density, high electron saturation velocity, and high temperature operation$^{11}$. Al$_x$Ga$_{1-x}$N/AlN heterostructure combines the photodetector abilities of deep ultra-violet (DUV) AlN along with the tunable bandgap Al$_x$Ga$_{1-x}$N, thereby also suppressing the visible spectrum and enhancing the UV/visible rejection ratio$^{12,13}$. This has applications in military target or missile detection, biochemical sensing, as solar-blind detectors, air/water purification, curing, and biomedical therapies and instrumentation$^{14-17}$. Considering the photodetector applications, AlN has a higher bandgap than Al$_x$Ga$_{1-x}$N and hence the resulting photodetector spectrum (of light waves incident on Al$_x$Ga$_{1-x}$N surface) would be dominantly dependent on the Al$_x$Ga$_{1-x}$N epitaxial layer properties; as opposed to other structures consisting of Al$_x$Ga$_{1-x}$N and a lower bandgap material, where it could be difficult to separate the effects of the two materials on the energy spectrum. Also, an Al$_x$Ga$_{1-x}$N/AlN structure would have the flexibility to function as a photodetector from top and bottom sides with front and back illumination respectively, with the top Al$_x$Ga$_{1-x}$N epitaxial layer having bandgap range from ~4 eV to ~6 eV depending on the Al content, and a bottom...
this is due to the lattice mismatch and thermal expansion difference between the thin films and substrates, which generally results in high-level strain-stress and mosaicity. Strain-stress varying within the epitaxial layers is one of the leading factors that reduces the electron mobility and degrades the device performance. Also, their optical and morphological properties could be improved by reducing the strain and stress. Therefore, it is vital to understand the strain and stress mechanism for improving the optical and electronic properties and applications of III-Nitrides.

High-resolution X-ray diffraction (HRXRD) and reciprocal space mapping (RSM) could be used to understand the crystal properties and to analyze the strain and stress in epitaxially grown III-Nitride films. The effect of different intermediate layers such as AlN, GaN, and step-graded AlGa1−xN/GaN HEMT structures on silicon (111) substrate has been studied by XRD, RSM and Hall effect measurements, showing that the in-plane stress can largely affect the two-dimensional electron gas mobility and carrier concentration.

The origin of stresses in AlGa1−xN/GaN heterostructures grown on c-plane sapphire substrate relies mainly on the thickness and growth temperature of the layers, alloy composition, device structure, and doping. In the case of AlGa1−xN/GaN superlattices, a compressive strain is observed in the GaN layers, and a tensile strain is observed in the AlGaN layers. The combination of these two strains could improve the crystal properties, dislocation densities, and coherence lengths are discussed. The effect of strain on the optical properties of the AlGa1−xN thin films has been investigated using photoluminescence (PL) and Raman spectroscopy.

Results and Discussion

The crystal structure and lattice parameters of MOCVD-grown AlGa1−xN and AlN have been studied using HRXRD and RSM techniques, while photoluminescence and Raman measurement results are discussed to understand the bandgap and phonon modes in AlGa1−xN and AlN. Figure 1 shows the 2θ–ω Bragg reflections around (0002) crystal planes for AlGa1−xN with varying x. The effect of strain is taken into account to determine the x values as per the synchrotron radiation HRXRD results. Bragg reflection peaks of (0002) from AlGa1−xN and AlN, and of (0006) from the sapphire substrate, are observed. The satellite peaks or the Laue oscillations in AlGa1−xN could be due to relatively smoother surface of AlGa1−xN with 35% Al or
due to the scattering of x-rays within the Al0.75Ga0.25N and the AlN layers. However, the primary goal here is to investigate the effect of Al content on the dominant and defining (0002) peak in the epitaxial layers.

The out-of-plane c-axis lattice constant (c) of Al0.75Ga0.25N thin films were calculated as shown in Table 1. Vegard’s law provides reliable unstrained lattice constants (c0, a0) for Al0.75Ga0.25N films using the bandgaps of GaN and AlN, and considering the very small lattice mismatch (~2%) between GaN and AlN. The calculated c, is lower than the unstrained c0, indicating a compressive strain along the c-axis (out-of-plane) in the Al0.75Ga0.25N thin films.

RSM based analysis were also done to determine the lattice constants and the stress-strain phenomenon in Al0.75Ga0.25N with changes in x. Figure 2 shows the symmetric plane RSM in the (0002) direction. A clear broadening of Al0.75Ga0.25N reciprocal lattice points (RLPs) reflection intensity distribution towards Qz, and Qx is seen. It can be observed that the maximum reflection intensity of Al0.75Ga0.25N shifts to higher Q values as the lattice constant c reduces, as x increases, which agrees very well with the results obtained from the 20–ω scan. Also, broadening along the Qz direction increases with x. Changes in the RSM plots with different Al content seem to be dominated by the Al0.75Ga0.25N layer.

Reciprocal space map around the AlN asymmetric (1013) RLP is illustrated in Figure 3. Based on the information from the asymmetric RSM scan, lattice parameters (a and c) were calculated for the hexagonal structure Eq. (1)37–39:

$$a = \frac{2\pi}{Q_x} \sqrt{\frac{4(h^2 + k^2 + h\ell)}{3}}, \quad c = \frac{2\pi l}{Q_Z},$$

(1)

Table 1 presents the calculated lattice parameters from the asymmetric RSM measurement (in this particular case, h = 1, k = 0, and l = 3) for Al0.75Ga0.25N. The calculated c from asymmetric RSMs is very close to the one obtained by HRXRD 20–ω scans for each sample, with a difference of about 0.06%; hence only the c-parameters from the HRXRD results are shown. The calculated a is larger than the unstrained one (a0) obtained by Vegard's law, which is due to the tensile strain along the a-axis (in-plane) in the Al0.75Ga0.25N epitaxial layers. Also, the a-lattice constant reduces with an increase in x, similar to c. A reduction in the lattice size and increase in the strain is seen in Al0.75Ga0.25N with an increase in the Al content in the alloy.

Figure 3 shows that with increasing Al composition, the maximum reflection intensity of Al0.75Ga0.25N RLPs progressively shifts from a partially relaxed (R = 1) towards a fully strained (R = 0) position. Since the AlN layer is thinner (~120 nm) than the Al0.75Ga0.25N layer (~800 nm), its reflection peak intensity is lower than Al0.75Ga0.25N. The intensity of Al0.75Ga0.25N RLPs broadens along the direction associated with the relaxation of the layer (the dashed black line). The Al0.75Ga0.25N RLPs get closer to the fully strained position with an increase in x. Note that both AlN and Al0.75Ga0.25N have a similar Q value of ~2.38 Å–1. An increase in Q value of ~2.38 Å–1 is observed with Al incorporation in Al0.75Ga0.25N, despite of reductions in lattice mismatch. As seen in Figure 3, a strain complementary to Al0.75Ga0.25N is induced in the AlN intermediate layer which increases with x as the Al0.75Ga0.25N layer is relaxed and adds to the inherent strain that is already present in AlN. The broadening in the symmetric and asymmetric RLPs implies an increase in the screw and edge dislocations (which are in the order of 109–1010 cm–2) respectively with x. The RSM and the 20–ω results show that the dislocations and the coherence lengths in Al0.75Ga0.25N/AlN change with x. Lattice constants of hexagonal AlN are typically smaller than GaN and hence, a reduction in the lateral correlation lengths and an increase in the dislocations are seen as Al0.75Ga0.25N composition is varied from GaN to AlN.

The overall in-plane strain (εa) and out-of-plane strain (εc) in the Al0.75Ga0.25N layers were determined using Eq. (2)38,40–42:

$$\varepsilon_a = \frac{a - a_0}{a_0}, \quad \varepsilon_c = \frac{c - c_0}{c_0},$$

(2)

The calculated strains (εa and εc) are attributed to the biaxial (εa and εc) and hydrostatic (εh) strains as shown in Eq. (3)34,36. (εa and εc) are the biaxial strains along a- and c-directions, respectively.

$$\varepsilon_a = \varepsilon_a^b + \varepsilon_a^h, \quad \varepsilon_c = \varepsilon_c^b + \varepsilon_c^h,$$

(3)

where εh is defined as $\varepsilon_h = \frac{1}{3} \epsilon_a (1 - \nu) (\varepsilon_a + \varepsilon_c)$, ν is Poisson’s ratio of Al0.75Ga0.25N calculated using Vegard’s law $\nu_{Al0.75Ga0.25N}(x) = x_{AlN} + (1 - x_{AlN})\nu_{AlN}$ and shown in Table 1. For the hexagonal crystal structure, the in-plane biaxial stress (σb) in the Al0.75Ga0.25N epitaxial layer can be determined by $\sigma_b = M_{12} \varepsilon_a + M_{13} \varepsilon_c$, where M12 is the biaxial elastic modulus given by $M_{12} = \frac{C_{11} + C_{12} + 2C_{13}}{C_{33}}$. The elastic constants (C0) of Al0.75Ga0.25N (Table 1) can be obtained by

| Al composition (x) | In-plane lattice parameter [Å] | Out-of-plane lattice parameter [Å] | Elastic constant [GPa] | Poisson ratio (v) |
|--------------------|---------------------------------|-----------------------------------|------------------------|-----------------|
| x = 0.23           | Calculated (a) Unstrained (a0) | Calculated (c) Unstrained (c0) | $C_{11}$, $C_{12}$, $C_{13}$, $C_{33}$ | $v$ |
| x = 0.47           | 3.190                           | 5.121                             | 3.138                  | 3.169 | 3.152 | 5.061 | 5.088 | 399.87 | 146.88 | 102.71 | 393.77 | 0.207 |
| x = 0.75           | 3.169                           | 4.998                             | 3.031                  | 405.75 | 148.00 | 100.75 | 391.25 | 0.205 |
Vegard’s law \( (C_{ij}^{\text{AlN}} - C_{ij}^{\text{GaN}} = x (C_{ij}^{\text{AlN}}) + (1-x) (C_{ij}^{\text{GaN}}) \) \). The calculated strains, biaxial strains, hydrostatic strain, and biaxial stress for Al\(_{x}\)Ga\(_{1-x}\)N epitaxial layers are summarized in Table 2. It can be seen that the in-plane (biaxial) strains are tensile, while the out-of-plane (biaxial) strains are compressive because of the different lattice mismatch along the in-plane and out-of-plane axes\(^{19}\) as also seen in the HRXRD results.

The biaxial strain has values close to the total strain in Al\(_{x}\)Ga\(_{1-x}\)N due to the relatively smaller values of \( \varepsilon_h \) and very few impurities introduced during growth. Also, the full width at half maximum (FWHM) values of the HRXRD (0002) \( \omega \) scans (not shown here) are found to be 627, 642, and 847 arcsec for Al\(_{0.23}\)Ga\(_{0.77}\)N, Al\(_{0.47}\)Ga\(_{0.53}\)N, and Al\(_{0.75}\)Ga\(_{0.25}\)N, respectively (Table 3)\(^{32}\). The lateral coherence lengths would range from 100 nm to 200 nm and have inverse proportionality with the Al content, indicating that the Al\(_{x}\)Ga\(_{1-x}\)N samples used in this study are of good crystal quality.

The broadening of the FWHM of (0002) HRXRD \( \omega \) scans in Al\(_{x}\)Ga\(_{1-x}\)N could be associated with the screw (c-type) threading dislocation (TD) along the c-axis. Figure 4(a) presents the compositional dependence of screw (c-type) TD density and out-of-plane strain in the Al\(_{x}\)Ga\(_{1-x}\)N thin films. The dislocation density of the Al\(_{x}\)Ga\(_{1-x}\)N thin films can be estimated from:

\[
D_{\text{screw}} = \frac{\beta_0(002)^2}{4.35b_{\text{screw}}^2},
\]

where \( D_{\text{screw}} \) is the screw type TD\(^{24}\), \( \beta_0 \) is the FWHM of the (0002) \( \omega \) scan, and \( b_{\text{screw}} = 5.1855 \) Å is the Burgers vector length for screw-type TD. As \( x \) increases, both the screw type TD density and the strain increase (Fig. 4(a)).
Evidently, the high density of screw dislocation observed in the Al-rich samples originated from a compressive strain along the $c$-axis (up to 0.6%) and a biaxial stress (up to 6.313 GPa), in $\text{Al}_x\text{Ga}_{1-x}\text{N}$, as presented in Table 3.

Photoluminescence measurements (Figure 4(b)) further indicate and help to understand the strain and stress in the epitaxial layers. A broadening of the $\text{Al}_x\text{Ga}_{1-x}\text{N}$ peaks is observed with an increase in $x$. Also, there is a shift in the peak positions compared to the unstrained energy gaps that are predicted by Vegard’s law. The PL peak positions are measured at 3.88, 4.27, and 5.25 eV for $\text{Al}_{0.23}\text{Ga}_{0.77}\text{N}$, $\text{Al}_{0.47}\text{Ga}_{0.53}\text{N}$, and $\text{Al}_{0.75}\text{Ga}_{0.25}\text{N}$, respectively. According to Vegard’s law, the predicted energy gap values for $x = 0.23, 0.47, \text{and} 0.75$ are 4.06, 4.73, and 5.51 eV respectively ($E_g(\text{AlN}) = 6.2 \text{ eV}$, $E_g(\text{GaN}) = 3.42 \text{ eV}$). If a bowing parameter of 1 eV is taken into consideration, the predicted bandgap values are 3.88, 4.47, and 5.32 eV for $x = 0.23, 0.47, \text{and} 0.75$, respectively. Smaller bandgap in the measured samples as compared to the predicted values, could be attributed more to the stronger tensile strain effect along the $a$-axis direction than the $c$-axis compressive strain ($\varepsilon_a \approx 2\varepsilon_c$) in the $\text{Al}_x\text{Ga}_{1-x}\text{N}$ epitaxial layers and hence, to the overall larger lattice constants of $\text{Al}_x\text{Ga}_{1-x}\text{N}$ epitaxial layers as compared to unstrained $\text{Al}_x\text{Ga}_{1-x}\text{N}$. The difference between the predicted and measured bandgap values is more for $x = 0.47$ and 0.75 than $x = 0.23$ due to more residual strain in $\text{Al}_x\text{Ga}_{1-x}\text{N}$ with high Al composition. Also, the bandgap increases with $x$ as would be expected and seems to be tunable between GaN and AlN. The PL peak broadening, intensity suppression and peak shifts could have multiple origins such as a statistical variation in the composition, Al-induced alloy disorder, strain and dislocations.

Raman spectra of the $\text{Al}_x\text{Ga}_{1-x}\text{N}$ samples under 532 nm excitation are shown in Figure 5. Two-mode behavior for the $E_{2}^{\text{high}}$ phonon$^{77}$ and one-mode behavior for the $A_{1}^{\text{LO}}$ phonon$^{89}$ are seen. Here, $E_{2}^{\text{high}}$ and $A_{1}^{\text{LO}}$ phonon modes correspond to the atomic oscillations in the $c$-plane (parallel to the $a$-axis) and along the $c$-axis, respectively. The phonon peaks exhibit a shift with increasing $x$. The $E_{2}^{\text{high}}$ (GaN-like) phonon is located at 575, 587, and ...

| Al composition ($x$) | FWHM of HRXRD [arcsec] | Screw TD Density [cm$^{-2}$] | FWHM of PL [meV] | Energy gap [eV] |
|----------------------|-------------------------|----------------------------|------------------|----------------|
| $x = 0.23$           | 627                     | $7.9 \times 10^8$          | 74               | 3.88           |
| $x = 0.47$           | 642                     | $8.3 \times 10^8$          | 100              | 4.27           |
| $x = 0.75$           | 847                     | $1.4 \times 10^9$          | 206              | 5.25           |

Table 3. Summary of structural and optical results of the $\text{Al}_x\text{Ga}_{1-x}\text{N}$ thin films.

Figure 4. Compositional dependence of (a) screw (c-type) TD density and out-of-plane strain, (b) PL FWHM and in-plane strain of $\text{Al}_x\text{Ga}_{1-x}\text{N}$ layers. The inset shows the room temperature PL spectra of $\text{Al}_{0.23}\text{Ga}_{0.77}\text{N}$, $\text{Al}_{0.47}\text{Ga}_{0.53}\text{N}$, and $\text{Al}_{0.75}\text{Ga}_{0.25}\text{N}$.
buffer layer. Finally, a ~800 nm Al substrate at 600 °C. The temperature was then increased to 1040 °C to grow a ~100 nm high-temperature (HT) AlN alloying. Considering the potential of Al

The strain increases with...compositions.

Figure 5. Raman spectra for AlxGa1−xN/AlN thin films (x = 0.23, 0.47, 0.75) measured with a 532 nm excitation laser under ambient conditions. The Raman spectra for x = 0.47 and 0.75 are multiplied by a factor of two for clarity. The dashed lines marking the composition dependence of the E2,high (GaN-like) and A1,low modes are guides to the eye. Asterisks near 576 (only observable for x = 0.75 because of overlapping with the E2,high (GaN-like) mode) and 750 cm−1 show the c-plane sapphire substrate phonons.

607 cm−1 for x = 0.23, 0.47, and 0.75, respectively, while the E2,high (AlN-like) phonon is located at ~650 cm−1 with a weak composition dependence. The A1,low phonon also exhibits strong composition dependence, from 783 to 864 cm−1 when x increases from 0.23 to 0.75. A sharp peak at 750 cm−1 (marked with an asterisk) and a weak peak at 576 cm−1 (marked with an asterisk and most visible for x = 0.75 because the peak is overlaid by the strong E2,high (GaN-like) peak) correspond to phonon vibrations of the the sapphire substrate. The composition-dependence behavior of the E2,high (GaN-like) and A1,low modes is in good agreement with previous work on AlxGa1−xN epilayers wherein the Raman results also confirm the wurtzite structure of the AlxGa1−xN layer with its hexagonal [0001] crystal plane parallel to the c-plane sapphire substrate. Strain due to alloying seems to be the major mechanism for the observed Raman shifts (the difference in phonon energies due to substrate-induced strain is small). Moreover, the E2,high (AlN-like) peak intensity varies with x, as the phonon vibrations are sensitive to atom compositions. Therefore, higher x values revealed more distinct E2,high (AlN-like) phonon vibration peaks, which is typical of alloy semiconductors. The result also suggests that the AlN buffer layer quality is good, so there is a small substrate-induced strain in the AlxGa1−xN epilayer.

Conclusion

In summary, the study focuses on the strain-stress status of AlxGa1−xN epitaxial layer grown by MOCVD on a c-plane sapphire substrate with AlN as intermediate layers. The lattice parameters reduce as the Al content in AlxGa1−xN is increased. The out-of-plane strain of AlxGa1−xN is compressive, and the in-plane strain is tensile. The strain increases with x, even though the lattice mismatch between AlxGa1−xN and AlN reduces. Broadening of the RSM peaks and the HRXRD rocking curve scans imply a consistent reduction in correlation lengths and higher dislocation densities with increasing x as the AlxGa1−xN composition is varied from GaN to AlN. The bandgap of AlxGa1−xN increases with x, as expected. Also, the values are smaller than the unstrained bandgap predicted by Vegard’s law, due to a larger tensile strain on the a-axis compared to the compressive strain on the c-axis. The E2,high and LO phonons exhibit a shift with an increasing x caused due to the strain accompanied with alloying. Considering the potential of AlxGa1−xN for optical and electronic applications, this work adds towards the understanding of crystal and optical properties of AlxGa1−x1−xN/AlN epitaxial layers.

Methods

Metal-organic chemical vapor deposition (MOCVD) growth. AlxGa1−xN thin films with varying x were grown on c-plane sapphire substrates by metal-organic chemical vapor deposition (MOCVD). The precursors for Al, Ga, and N, are trimethylaluminum (TMA), trimethylgallium (TMG), and ammonia (NH3), respectively. To remove surface contamination, sapphire substrates were heated at 1100 °C in H2 ambient prior to the growth. A 40 Torr chamber pressure was maintained for the growth of AlN and AlxGa1−xN epitaxial layers. A ~20 nm low-temperature (LT) AlN nucleation layer with a V/III ratio of 3000 was deposited on the sapphire substrate at 600 °C. The temperature was then increased to 1040 °C to grow a ~100 nm high-temperature (HT) AlN buffer layer. Finally, a ~800 nm AlxGa1−xN epitaxial layer was grown on the AlN layer at 1140 °C. The samples were cooled in NH3 environment.

Materials characterization. Synchrotron radiation HRXRD measurement were performed at 33IDD beamline at the Advanced Photon Source, Argonne National Laboratory. It is equipped with a standard six-circle Kappa-type diffractometer and Pilatus 100 K area detector. A deep ultraviolet (DUV) PL spectroscopy (excitation...
at 224 nm) was used to measure the optical properties of the Al\(_{0.5}\)Ga\(_{0.5}\)N thin films. Micro-Raman spectroscopy was performed using a Horiba Jobin-Yvon XploRA confocal Raman spectrometer in a backscattering configuration with a 532 nm excitation laser and a grating of 1800 lines/mm.

**Data Availability**
The datasets generated during and/or analyzed in the current study are available from the corresponding author on reasonable request.

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Acknowledgements
The authors at Purdue University are grateful for the financial supports from National Science Foundation CAREER program (under Grants of CMMI - 1560834) and NSF IIP- 1700628. The authors thank Professor Yong P. Chen at Purdue University for allowing the use of Raman spectroscopy equipment in his lab. This research used resources of the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357.

Author Contributions
N.L. and I.F. contributed to the conception and design of experiments. Y.F., V.S., T.C. and B.K., conducted most of experiments, characterization and drafted the manuscript. Y.D. and H.Z. conducted synchrotron and reciprocal space mapping. N.L., I.F. and H.Z. edited and revised the manuscript. N.L. and I.F. supervised the project. All the authors discussed the results.

Additional Information
Competing Interests: The authors declare no competing interests.

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