



Title	Metalorganic Vapor-Phase Epitaxy of +c/-c GaN Polarity Inverted Bilayer for Transverse Quasi-Phase-Matched Wavelength Conversion Device
Author(s)	Ikeda, Kazuhisa; Malik, Shahzeb; Uemukai, Masahiro et al.
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Metalorganic Vapor-Phase Epitaxy of $+c/-c$ GaN Polarity Inverted Bilayer for Transverse Quasi-Phase-Matched Wavelength Conversion Device

Kazuhisa Ikeda,* Shahzeb Malik, Masahiro Uemukai, Tomoyuki Tanikawa,* and Ryuji Katayama

Photon-pair generation based on optical parametric down-conversion has attracted for the application as a light source for quantum information. Highly efficient wavelength-conversion devices require a polarity-inversion structure when using nitride semiconductors. A transverse quasi-phase-matching (QPM) polarity-inverted GaN bilayer channel waveguide device is suitable for efficient wavelength conversion. This study designed a cross-section device to satisfy the modal dispersion phase-matching condition between the TM_{02} mode pump light and the TM_{00} mode signal/idler light. Moreover, an AlN oxidation interlayer fabricates the Ga-polar/N-polar ($+c/-c$) GaN layers via metalorganic vapor-phase epitaxy (MOVPE). A 145 nm thick film layer with a macro-step-free surface is grown by optimizing the $-c$ -GaN growth conditions and reducing the substrate off-angle to 0.2° . Next, the AlN layer is oxidized in an electric furnace and MOVPE is used to regrow a 1500 nm thick $+c$ -GaN layer. A macrosteps-free surface can be achieved by reducing the off-angle to 0.2° and optimizing the $-c$ -GaN growth conditions to avoid hillock formation. These results pave the way for improving the efficiency of GaN transverse QPM wavelength-conversion devices.

1. Introduction

Nitride semiconductors, including AlN, GaN, and InN, and their alloys are suitable for blue light-emitting diodes (LEDs),^[1,2] ultra-violet LEDs,^[3–5] and laser diodes.^[6] Nitride semiconductors possess a wurtzite structure that exhibits strong polarization along the c -axis direction^[7] and are applicable in wavelength-conversion devices, such as ultraviolet second-harmonic generation (SHG) devices and quantum light sources based on optical parametric

down-conversion (OPDC).^[8] Nitride semiconductors, being paraelectric materials, require the fabrication of periodically polarity-inverted structures via polarity-controlled heteroepitaxial growth, and this is necessary to satisfy the quasi-phase-matching (QPM) condition between the pump and converted light. Conventional nitride-semiconductor-based wavelength-conversion devices have a longitudinal QPM structure where the polarity is periodically inverted along the light propagation direction. The longitudinal QPM structures were fabricated by simultaneous growth of metal-polar ($+c$) and N-polar ($-c$) domains.^[9] However, there are variations in the rate at which atoms are incorporated locally, depending on the growth polarity. When the growth rate of the $-c$ -GaN domains surpassed that of the $+c$ -GaN domains, lateral overgrowth of the $-c$ -GaN regions over the $+c$ -GaN regions

was observed,^[10] resulting in enhanced surface scattering of light.^[11,12] Wavelength conversion can be accomplished by utilizing a transverse quasi-phase-matching QPM waveguide. This waveguide is designed to satisfy the modal dispersion phase-matching (MDPM) condition between the pump and converted light with different modes.^[13,14] The mismatch in the mode order of the pump and converted light reduces the nonlinear coupling coefficient κ , hence limiting the wavelength-conversion efficiency.^[15] The transverse QPM waveguide with a polarity-inverted structure perpendicular to the light propagation direction is used to increase the κ value, in which the polarity-inversion interface is located at the node of the high-order mode electric field.^[16–20]

Polarity-inverted stacked bilayers of GaN can be fabricated by using the following approaches: thin-film transfer by utilizing surface-activated bonding^[20] and polarity inversion via epitaxial growth. The polarity of the films can be inverted by incorporating a heavily Mg-doped layer during the growth of GaN,^[21,22] or by inserting an AlN oxidation layer during the growth of AlN.^[23] One study also reported that the AlN oxidation layer can be used as an interlayer for epitaxial polarity inversion of GaN. Wong et al. showed the polarity inversion of GaN films from $-c$ to $+c$ using plasma-assisted molecular beam epitaxy (PAMBE) by introducing a thin AlN/ AlO_x interlayer.^[24] Moreover, Hite et al.

K. Ikeda, S. Malik, M. Uemukai, T. Tanikawa, R. Katayama
Graduate School of Engineering
Osaka University
2-1, Yamadaoka, Suita, Osaka 565-0871, Japan
E-mail: ikeda.k@qoe.eei.eng.osaka-u.ac.jp;
tanikawa@eei.eng.osaka-u.ac.jp

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demonstrated polarity inversion of GaN films from $-c$ to $+c$ using metal-organic vapor-phase epitaxy (MOVPE) and this was achieved by inserting a low-temperature, thin interlayer of either AlN^[25] or an Al₂O₃.^[26] The epitaxial polarity-inversion method is most favorable due to its fewer process steps as compared to the thin-film transfer method. The bilayer thickness in a transverse QPM waveguide is ≈ 500 nm to satisfy the MPDM condition. Therefore, it is necessary to employ heteroepitaxial growth with submicron-scale thickness and epitaxial inversion techniques to obtain a smooth surface, which is essential for low-optical-loss transverse QPM devices. $-c$ -GaN is a suitable bottom layer over the sapphire substrate because it allows for heteroepitaxial growth with a two-dimensional growth mode at an earlier stage compared to the growth of $+c$ -GaN.^[27] In our previous study, we successfully demonstrated the growth of a $+c$ -GaN/ $-c$ -GaN bilayer with an AlN oxidation layer by MOVPE. To avoid the formation of hillocks during the growth of $-c$ -GaN, we utilized a 0.8° off-oriented sapphire substrate.^[28] However, macrosteps were observed over the entire $-c$ and regrown GaN surfaces due to the step-bunching phenomenon.^[29,30]

This study demonstrated the growth of thin $-c$ -GaN films on 0.2° off sapphire substrates by optimizing the growth conditions, including the sapphire nitridation time and buffer layer thickness. The aim was to achieve a smooth surface without any macrosteps or hillocks, with a thickness in a submicron range. Subsequently, we fabricated a $+c/-c$ GaN polarity-inverted channel waveguide by employing epitaxial polarity inversion, photolithography, and dry etching techniques.

2. Device Design and Epitaxial Polarity Inversion Process

A schematic of the GaN transverse QPM channel waveguide is illustrated in **Figure 1**. The channel waveguide comprises a $+c/-c$ polarity stacking bilayer surrounded by a sapphire substrate and a SiO₂ cladding layer. To fabricate the waveguide structure, the electric field distributions, and effective refractive indices were computed using the finite-difference approach.^[31] Using real samples, the refractive-index dispersions of SiO₂, GaN, and sapphire were measured by a spectroscopic ellipsometer. The channel waveguide thickness was 436 nm for a channel width of $1.0 \mu\text{m}$ when the MDPM condition between the pump wave (TM₀₂ guided mode at 405 nm wavelength) and the signal and idler waves (TM₀₀ guided modes at 810 nm wavelength) was satisfied. The κ value can be increased by introducing polarity inversion located at the node of the TM₀₂ electric field. κ was

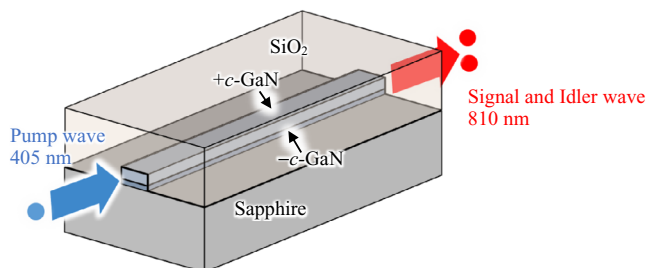


Figure 1. Schematic of GaN transverse QPM channel waveguide.

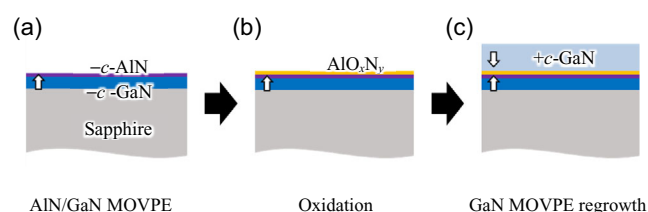


Figure 2. Fabrication process of polarity-inverted GaN bilayer using MOVPE. a) AlN/GaN MOVPE, b) oxidation, and c) GaN MOVPE regrowth.

calculated to be $2.18 \text{ W}^{-1/2} \text{ cm}^{-1}$ when the polarity-inverted interface is situated at the lower node of the TM₀₂ electric field, and the thicknesses of the lower and top GaN layers are 145 and 291 nm, respectively.

Figure 2 shows the proposed epitaxial inversion method utilizing MOVPE. a) The initial stage involved the growth of a 145 nm thick $-c$ -GaN film and a 10 nm thick AlN film on a nitrided sapphire substrate by using MOVPE, b) then the sample was subjected to annealing in an oxygen atmosphere using an electric furnace. c) MOVPE was used again to grow a $2.1 \mu\text{m}$ thick $+c$ -GaN film on top of the AlN oxidation film. Due to the inability of three-dimensional growth to cover the surface of $+c$ -GaN growth at the early stage, the growth process proceeded until a film thickness of $2 \mu\text{m}$ was reached, which was adequate for two-dimensional growth. The detailed growth conditions are described in the Experimental Section. To obtain the thickness for the fabrication of the channel waveguide that satisfies the MDPM condition, an etch-down process is required. The incorporation of the 10 nm thick AlN oxidation layer slightly shifts the phase-matching wavelength from the designed value owing to the structural differences. The presence of a high density of oxygen impurities in the AlN layer does not have any impact on the absorption properties of light at both 405 and 810 nm. Slack et al. have determined that the oxygen absorption region in the AlN film lies between 3.5 and 5.2 eV.^[32] Another concern arises from the decrease in κ value due to the change in the distribution of the nonlinear optical constants inside the core region. Nevertheless, its effect is expected to be minimal due to the very thin AlN oxidation layer located at the node of a high-order electric field profile, resulting in a negligible impact on nonlinear optical coupling.

A suitable surface roughness value was initially determined because surface roughness can result in loss due to scattering. The Payne–Lacey model is a commonly used method for describing the scattering loss in waveguides as a function of fundamental waveguide parameters and root-mean-square (RMS) values.^[33,34] The scattering loss is determined by the given equation

$$\alpha = 2.17 \frac{\sigma^2}{\sqrt{2} k_0 d^4 n_1} g f \quad (1)$$

where α represents the waveguide scattering loss, σ^2 , k_0 , d , and n_1 are the mean square deviation of the surface roughness, free-space wave vector, waveguide half-height, and core index, respectively. The function g is defined by the waveguide geometry, whereas f is determined by the index step of the waveguide. When the RMS value decreases to 100, 10, and 1 nm, the waveguide scattering loss will be 3.4×10^2 , 3.4, and $3.4 \times 10^{-2} \text{ cm}^{-1}$,

respectively. The objective of this study is to achieve an RMS value in the single digits to minimize waveguide loss.

3. Results and Discussion

3.1. Heteroepitaxy of Thin $-c$ -GaN Films on Sapphire Substrates

Initially, we examined how the duration of nitridation over the sapphire substrate impacted the surface morphology of the $-c$ -GaN film. **Figure 3** displays the $700 \times 700 \mu\text{m}^2$ white-light interferometry images of the 145 nm thick $-c$ -GaN films. This figure corresponds to a sapphire nitridation period that ranges from 10 to 30 min. When the sapphire nitridation time is only 10 min, the film exhibits insignificant grains with a small domain size. These islands will result in significant scattering loss; hence, large grains are preferable. Upon increasing the duration of surface nitridation to 15, 20, and 30 min, hexagonal hillocks with an average size of $40 \mu\text{m}$ were found. The hillock densities in the 145 nm thick $-c$ -GaN films were 160, 24.5, and 49.0 cm^{-2} for the nitridation durations of 15, 20, and 30 min, respectively. The RMS roughness values of the 145 nm thick $-c$ -GaN films were measured to be 76.5, 29.5, and 39.6 nm for nitridation times of 15, 20, and 30 min, respectively. After a nitridation time of 10 min, we observed numerous insignificant grains. The density of the hillocks decreased once the nitridation time increased to 20 min. However, when the duration of the nitridation was increased to 30 min, an increase in hillock density was observed. These trends were attributed to changes in the sapphire surface conditions caused by differences in nitridation

time. Stolyarchuk et al. assessed the effects of the nitridation time on the surface morphology.^[35] Hillock formation can occur on substrates with low off-angle.^[36] The main sources of hillocks were spiral growth that originated from the inversion domain boundaries (IDBs) formed at the film–substrate interface,^[37–39] as well as the screw threading dislocations.^[40,41] The growth polarity depended on the nitridation process parameters, particularly the temperature and time. When the sapphire nitridation was performed over 1000°C , $+c$ -AlN islands formed at the GaN–sapphire interface.^[42] This triggered the formation of IDBs and hillock growth in the subsequent GaN epilayer. In this work, limiting the nitridation time to 10 min resulted in the formation of the $\text{Al}_x\text{O}_y\text{N}_z$, which in turn led to $+c$ -polarity IDBs.^[43] However, when the nitridation time was extended to 30 min, excessive nitridation occurred, leading to the development of $+c$ -AlN crystals on the sapphire substrate and consequently resulting in the formation of IDBs in the $-c$ -GaN epilayer.^[37] Therefore, this study determined an ideal surface nitridation time of 20 min. However, the RMS roughness value remained significant for fabricating a transverse QPM channel waveguide with a submicron-order thickness.

In addition, the effect of the growing process of the low-temperature GaN (LT-GaN) buffer layer growth process was investigated. **Figure 4** shows the white-light interferometry images of the $-c$ -GaN films. These films were grown using a trimethylgallium (TMG) flow rate of either 20 or $24 \mu\text{mol min}^{-1}$, and a growth time of either 135 or 180 s during the LT-GaN buffer layer growing process. The LT-GaN buffer layer thicknesses in Figure 4a–d are expected to be 16, 19, 21, and 24 nm, respectively. When the TMG flow and growth time during the LT-GaN buffer layer growth were $20 \mu\text{mol min}^{-1}$ and

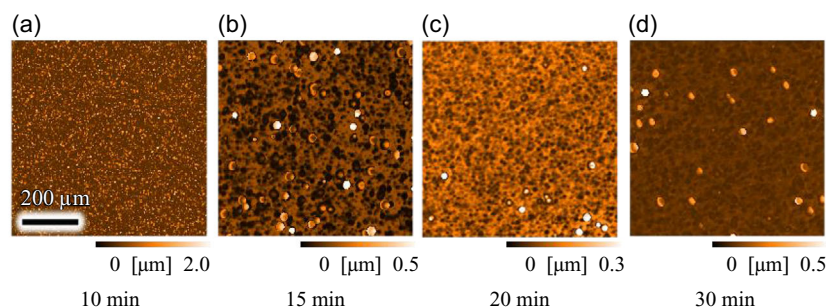


Figure 3. White-light interferometry images of the $-c$ -GaN films grown with different nitridation times. a) 10 min b) 15 min c) 20 min d) 30 min.

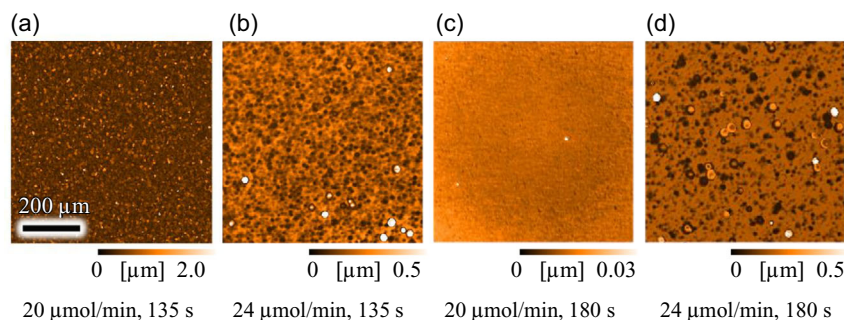


Figure 4. White-light interferometry images of $-c$ -GaN films at different TMG flows and times during the LT-GaN buffer layer growth. a) $20 \mu\text{mol min}^{-1}$, 135 s b) $24 \mu\text{mol min}^{-1}$, 135 s c) $20 \mu\text{mol min}^{-1}$, 180 s d) $24 \mu\text{mol min}^{-1}$, 180 s.

135 s, respectively, numerous fine hillocks appeared, resulting in a rough surface. Under different conditions, hexagonal hillocks with an average size of 40 μm were observed, and the hillock density of the 145 nm thick $-c$ -GaN films was found to be 24.5, 0.0, and 38.8 cm^{-2} , as shown in Figure 4b–d, respectively. The RMS values of the 145 nm thick $-c$ -GaN films were 29.5, 5.6, and 50.5 nm, as shown in Figure 4b–d, respectively. When the flow rate of TMG was 20 $\mu\text{mol min}^{-1}$, no hillocks were observed during the growth of the LT-GaN buffer layer, irrespective of the time variation from 135 to 180 s. Moreover, when the TMG flow rate was set at 24 $\mu\text{mol min}^{-1}$, hillock density remained unchanged for a duration of 135 s. However, hillocks were seen when the TMG flow rate was increased from 20 to 24 $\mu\text{mol min}^{-1}$ for 180 s during the growth process. This result has affected both the size and density of the growth nuclei of the LT-GaN buffer layer.^[44,45] It was expected that the thickness of the LT-GaN buffer layer would increase due to the increase in the TMG flow rate and growth time. When the LT-GaN buffer layer is too thin, the growth nuclei become insignificant and dense, resulting in a high screw dislocation density that triggers high-density hillock growth. On the contrary, if the LT-GaN buffer layer is too thick, the growth nuclei density decreases, but the size variation increases, resulting in the formation of large hillocks. Thus, the LT-GaN buffer layer fabricated for 180 s with a TMG flow of 20 $\mu\text{mol min}^{-1}$ is the most optimal for achieving the desired size and density of growth nuclei.

Figure 5 shows the AFM image of the $-c$ -GaN film grown over the LT-GaN buffer layer. The RMS roughness was 0.6 nm. Therefore, it was determined that a surface nitridation time of 20 min and the growth of the LT-GaN buffer layer for 180 s yielded optimal results. By optimizing the growth conditions, it was possible to fabricate a flat $-c$ -GaN film on the sapphire substrate with a low off-angle of 0.2°.

X-Ray diffraction (XRD) measurements were employed to evaluate the structural property of the 145 nm thick $-c$ -GaN film. Figure 6a shows the XRD 2θ - ω scan of the $-c$ -GaN film around the symmetric 0002 diffractions along with the simulation curve. The experimental profile reached its highest point at 34.55°, showing Pendellösung fringes, indicating that the thin film of GaN exhibited a smooth surface morphology. The period of the Pendellösung fringe confirms that the thickness of the $-c$ -GaN film was precisely 145 nm, as designed. Figure 6b,c display the X-Ray rocking curves (XRCs) for the symmetric 0002 and skewed symmetric 10 $\bar{1}2$ reflections of GaN; the full widths at half maximum (FWHMs) for these reflections were 11 and 1023 arcsec, respectively. The 0002 XRC spectrum contained both sharp and broad components. Similar profiles were obtained

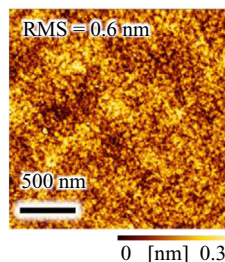


Figure 5. $2 \times 2 \mu\text{m}^2$ AFM image of $-c$ -GaN film.

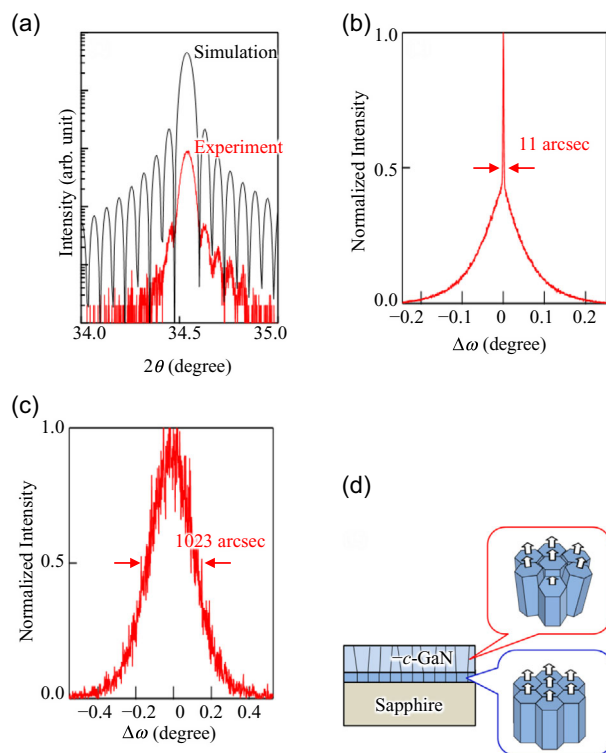


Figure 6. XRD measurement results of 145 nm thick $-c$ -GaN film. a) Symmetric 2θ - ω scan around 0002 diffraction. XRCs for b) symmetric 0002 and c) 10 $\bar{1}2$ reflections. d) Schematic illustrations of designed film.

for thin-sputtered AlN films on sapphire substrates, and their origins have been discussed in detail.^[46,47] The 0002 XRC profile reflects the presence of tilt mosaicity. A thin heteroepitaxial film was grown quasi-coherently on the substrate, and the 0002 XRC results showed only a sharp single peak. When the film thickness exceeded the relaxation point, a broad peak was observed, indicating the deterioration of the tilt mosaics. The thickness dependence of the 0002 XRC profile suggests the presence of a quasi-coherent layer at the bottom and a relaxing layer at the top, as illustrated in Figure 6d. Therefore, the unique 0002 XRC profile can be attributed to thin heteroepitaxial growth that has reached the strain relaxation point, as depicted in Figure 6b. The presence of both sharp and broad peaks in the GaN film is due to the pure edge dislocation networks, leading to inhomogeneous in-plane strain relaxation.^[48–50] The presence of high-density edge threading dislocations and strain relaxation increased the FWHM of the 10 $\bar{1}2$ reflection, as shown in Figure 6c.

3.2. Epitaxial Polarity-Inverted Structure Fabrication

An epitaxial polarity-inverted structure was fabricated by regrowing GaN films on the $-c$ -AlN/GaN layers using MOVPE. Figure 7 shows the in-situ reflectance traces of 950 nm light and pyrometer temperature transients during GaN regrowth. When the process of GaN regrowth was performed on the

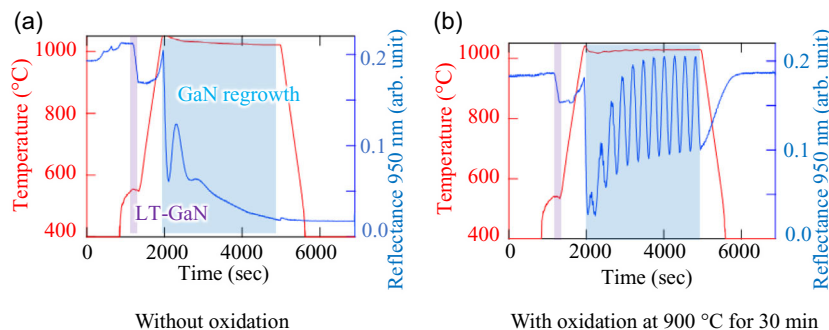


Figure 7. In-situ reflectance traces of 950 nm and pyrometer temperature transients for regrowth of GaN a) without and b) with oxidation at 900 °C for 30 min.

–*c*-AlN/GaN film without oxidation, the reflectance showed oscillations due to the interference light reflected from the GaN surface and the GaN–substrate interface. However, it reduced monotonically, indicating a notable increase in surface roughness. When the –*c*-AlN/GaN film was annealed at a temperature of 900 °C for 30 min, in-situ reflectance dropped during the initial GaN growth stage and thereafter increased with oscillation. The observed reflectance transient for +*c*-GaN growth on sapphire substrates^[51,52] indicates that polarity inversion has occurred successfully. Surface white-light interferometry images of the regrown GaN films are shown in **Figure 8a,b**, one with oxidation and one without. Numerous hillocks were observed on the regrown GaN surface without oxidation, indicating that the regrowth layer was dominated by –*c* polarity. The hillock density significantly decreased during the process of GaN regrowth by oxidation. The absence of macrosteps in the surface morphology can be attributed to the low off-angle of the substrate. The hillock density was $6 \times 10^2 \text{ cm}^{-2}$. The RMS roughness in the $700 \times 700 \mu\text{m}^2$ area was measured to be 27.3 nm, based on the significant height of the hillocks. The RMS roughness, estimated from the hillock-free region, was found to be 6.4 nm. According to Equation (1), the loss in the area without hillocks was $\approx 1.4 \text{ cm}^{-1}$ for the waveguide applications. This value is comparable to the value found in standard GaN-based waveguides.^[53] The polarity of the regrown GaN layer was determined via KOH etching performed at a temperature of 60 °C for 10 min. The stability of the alkaline solution was strongly dependent on the polarity of the film; the –*c*-polar regions were roughened by etching, while the +*c*-polar regions

were not etched.^[54] After the KOH etching, the surface morphology of the regrown GaN film experienced a change without oxidation while remaining unchanged in the presence of oxidation. This indicates that the successful epitaxial polarity inversion from –*c* to +*c* was achieved during the GaN regrowth on the –*c*-AlN layer with oxidation.

Figure 9 shows the AFM image of the +*c*/–*c*-GaN film after undergoing oxidation at a temperature of 900 °C for 30 min. The surface exhibits bilayer atomic steps and has an RMS roughness of 0.2 nm. The surface also contains small pits located at the atomic step terminations which correspond to the surface termination of the screw and mixed threading dislocations. The pit density was $1.0 \times 10^9 \text{ cm}^{-2}$.

Secondary ion mass spectrometry (SIMS) depth profiles of the fabricated inverted-polarity structures can be seen in **Figure 10**. The oxygen concentration peak can be seen at the –*c*-GaN/+*c*-GaN interface because of the AlN oxidation interlayer. The oxygen concentration in the –*c*-GaN layer is greater than in the +*c*-GaN layer due to the high oxygen-incorporation efficiency in the –*c*-GaN layer.^[55,56] The oxygen concentration in the +*c*-GaN layer was initially high from the regrowth interface to a depth of 1.3 μm . However, it gradually decreased and became stable at $\approx 2.5 \times 10^{16} \text{ cm}^{-3}$. The oxygen-rich region is identified in **Figure 7b** via the in situ reflectance profile which corresponds to a three-dimensional growth of the +*c*-GaN region. The three-dimensional growth islands have $\{10\bar{1}1\}$ side facets,^[57] which incorporate oxygen impurities more readily as compared to the +*c*-plane. The concentrations of other impurities, including carbon (C), hydrogen (H), and silicon (Si) were also increased at the polarity-inversion interface and remained constant in

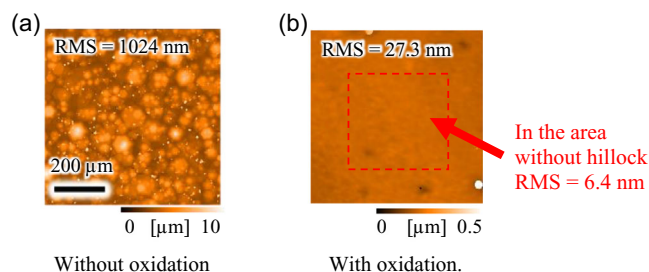


Figure 8. White-light interferometry images of regrown GaN films over –*c*-AlN/GaN films a) show the image without oxidation, whereas b) show the image with oxidation at 900 °C for 30 min.

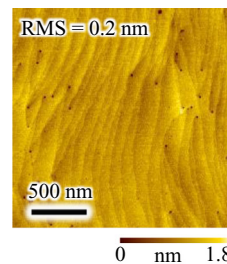


Figure 9. $2 \times 2 \mu\text{m}^2$ AFM image of +*c*/–*c*-GaN film with oxidation at 900 °C for 30 min.

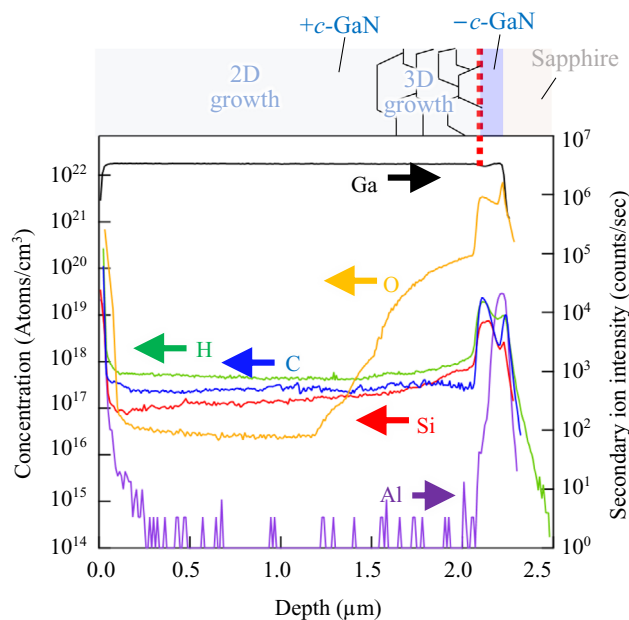


Figure 10. SIMS depth profiles for fabricated $+c/-c$ -GaN polarity-inversion stacked structure.

the $+c$ -GaN region. Sumi et al. reported the impact of oxygen concentration on the absorption properties of GaN substrates grown by oxide vapor-phase epitaxy.^[58] Absorption coefficients were $30\text{--}50\text{ cm}^{-1}$ for 810 nm and $50\text{--}100\text{ cm}^{-1}$ for 405 nm when the oxygen concentration was greater than $>10^{20}\text{ cm}^{-3}$ as shown in Figure 10. These high absorption coefficients in the channel waveguide result in propagation loss, which reduces the wavelength conversion efficiency. Further optimization of growth conditions should be taken into consideration to reduce absorption by oxygen impurities.

Figure 11 shows XRCs of symmetric 0002 and skew-symmetric $10\bar{1}2$ reflections of GaN for two different structures: the $-c$ -GaN film and the $+c$ -GaN/ $-c$ -GaN epitaxial polarity inverted structure. In the epitaxial polarity inverted structure, the top $+c$ -GaN film was much thicker ($2.1\text{ }\mu\text{m}$) than the bottom

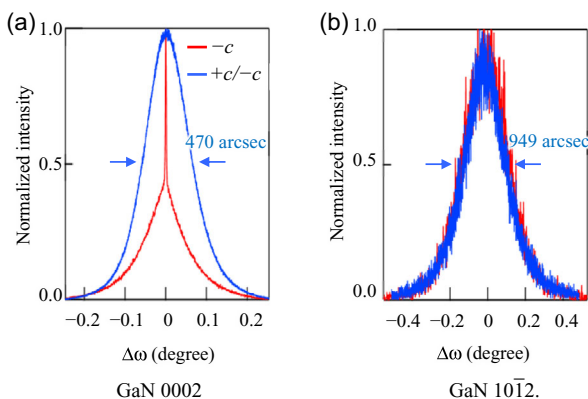


Figure 11. XRCs of $-c$ -GaN (red) and regrown $+c$ -GaN (blue) layers for a) 0002 and b) $10\bar{1}2$ reflections.

$-c$ -GaN layer, and as a result, the XRCs of the epitaxial polarity-inverted structure were mainly from the top $+c$ -GaN layer. Comparing the $-c$ -GaN layer to the $+c$ -GaN layer, the FWHM of the 0002 reflection increased from 11 to 470 arcsec, indicating an increase in the tilt mosaics. This increase is caused by additional threading dislocations due to strain relaxation from the oxidized AlN interlayer.^[28] Conversely, the FWHM for the $10\bar{1}2$ reflections of the $+c$ -GaN layer narrowed from 1023 to 949 arcsec, indicating a decrease of twist mosaics, and this decrease is attributed to the reduction in edge threading dislocation density during GaN growth, resulting from the annihilation and reaction of threading dislocations.^[59]

3.3. Channel Waveguide Fabrication

A wavelength-conversion device was fabricated using the $+c/-c$ -GaN polarity-inverted stacked structure. Figure 12 illustrates the fabrication process of the GaN transverse QPM channel waveguide. a) The $+c/-c$ -GaN polarity-inverted structure was fabricated by MOVPE. b) The top $+c$ -GaN layer was etched using inductively coupled plasma reactive-ion etching (ICP-RIE) until the remaining polarity-inverted GaN film reached a thickness of 436 nm. c) A 200 nm thick SiO_2 film was deposited via plasma-enhanced chemical vapor deposition (PECVD). d) A SiO_2 mask pattern was formed using maskless lithography, and e) SiO_2 mask was achieved by capacitively coupled reactive ion etching (CCP-RIE). f) The GaN channel waveguide was formed using ICP-RIE. g) Finally, a 700 nm thick SiO_2 cladding layer was deposited using the PECVD method and the end facets of the channel waveguide were formed by dicing and polishing.

To compare the change in surface roughness during the waveguide fabrication process, differential interference contrast microscope images of the $+c/-c$ -GaN film before (as-grown) and after ICP-RIE were compared as shown in Figure 13. The as-grown surface showed fewer hillock and pit densities and no bunched steps. The hillock and pit densities in Figure 13a are $\approx 1.2 \times 10^3$ and $1.8 \times 10^3\text{ cm}^{-2}$, respectively. While Figure 13b demonstrates that the hillocks and pits resulting from ICP-RIE lead to surface roughening, some regions maintain a smooth surface morphology. The hillocks and pits are possibly formed during the growth of $-c$ -GaN and $+c$ -GaN layers, respectively. Further optimization of growth conditions can be preferable to improve the yields of the process.

Figure 14 shows a cross-sectional scanning electron microscope (SEM) image obtained after polishing the end-facet. A polarity-inverted stacked-channel waveguide was successfully fabricated, achieving the required film thickness. The channel width exceeded the intended width of $1.0\text{ }\mu\text{m}$ and the end facet of the GaN channel waveguide has a trapezoidal shape. Because the effective refractive index of the guided mode was relatively insensitive to the waveguide width compared with the waveguide thickness, the deviation in the phase-matching wavelength was insignificant. The dark horizontal contrast at the polarization inversion interface indicated the presence of an AlN oxidation interlayer. The SEM image also shows that both the polarity inversion interface and the interface between the SiO_2 cladding layer and the $+c$ -GaN core were flat.

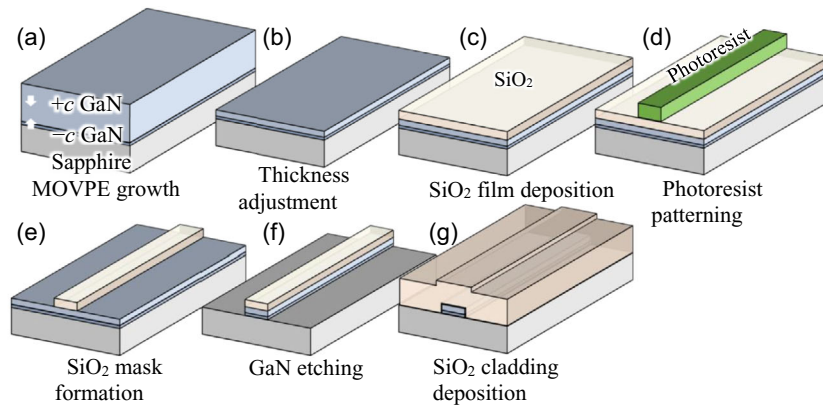


Figure 12. GaN transverse QPM channel waveguide fabrication process. a) MOVPE growth b) Thickness adjustment; c) SiO₂ film deposition; d) Photoresist patterning; e) SiO₂ mask formation; f) GaN etching; g) SiO₂ cladding deposition.

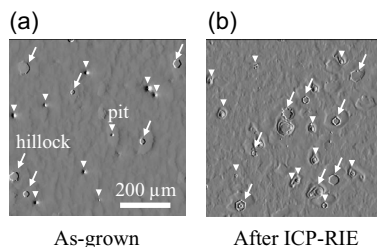


Figure 13. Differential interference contrast microscope images of +c/-c GaN film a) as-grown, b) after ICP-RIE. Hillocks and pits are indicated by arrows and triangles, respectively.

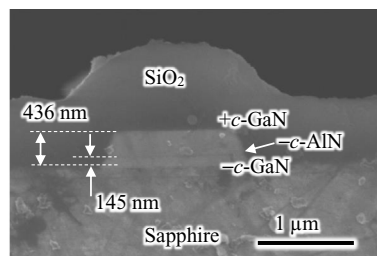


Figure 14. Cross-sectional SEM image of polished end facet of GaN channel waveguide.

4. Conclusion

This study involved the fabrication of a GaN polarity-inverted structure on a 0.2°-off sapphire substrate with a macrosteps-free surface. The fabrication process used MOVPE with an AlN oxidation interlayer. We investigated the most favorable parameters for fabricating a smooth surface of -c-AlN/GaN film on a 0.2°-off sapphire substrate. A sapphire surface nitridation time of 20 min and an LT-GaN buffer layer grown for 180 s with a TMG flow of 20 μmol min⁻¹ were required for achieving an -c-AlN/GaN film on a 0.2°-off sapphire substrate with a smooth surface over a 2 in. wafer. The growth of the GaN over the -c-AlN/GaN film with oxidation-enabled polarity inversion, and the surface exhibited no macrosteps and a lower hillock density. The results

demonstrated that MOVPE can effectively fabricate a transverse QPM wavelength-conversion device, as well as a transverse QPM channel waveguide can also be fabricated.

5. Experimental Section

The process of epitaxial growth was performed using an AIXTRON close-coupled showerhead (CCS) 3 × 2 MOVPE system. The MOVPE reactor was equipped with an in situ reflectance monitoring system (LayTec, EpiCurveTT), and *c*-plane sapphire substrates with an off angle of 0.2° toward the *m*-axis were used. TMG, trimethylaluminum (TMA), and ammonia (NH₃) were used as precursors for gallium, aluminum, and nitrogen, respectively. Hydrogen and nitrogen were used as the carrier gases. Initially, a process of hydrogen cleaning was performed at 1085 °C for 5 min. Afterward, NH₃ was introduced into the reactor to nitride the sapphire surface for 10–30 min. The LT-GaN buffer layer was grown at a temperature of 490 °C. The TMG flow rate and growth time for the LT-GaN buffer layer growth were either 20 or 24 μmol min⁻¹ and 135 or 180 s, respectively. A 145 nm thick -c-GaN layer was grown at a temperature of 1020 °C. The flows of TMG and NH₃ across the GaN growths were maintained constant at 149 μmol min⁻¹ and 3.0 slm, respectively. The V/III ratio for the -c-GaN growth was 900. In the epitaxial polarity-inversion process, a 10 nm thick AlN film with a TMA flow of 22.9 μmol min⁻¹ was grown over a 145 nm thick -c-GaN layer. The sample was loaded from the reactor and annealed at 900 °C in the presence of oxygen in an electric furnace to oxidize the AlN surface. Following that, the GaN film was regrown using MOVPE. Hydrogen may cause desorption from the oxide surface; the regrowth process was initiated using a nitrogen carrier gas. After increasing the temperature to 550 °C, a 16 nm thick LT-GaN buffer layer was grown at 540 °C using nitrogen carrier gas with a TMG flow rate of 24 μmol min⁻¹ and NH₃ flow of 2.0 slm. Following this, the temperature was elevated to 1020 °C and the carrier gas was changed to hydrogen. A 2.1 μm thick GaN layer was then grown at 1020 °C using a TMG flow of 149 μmol min⁻¹ and an NH₃ flow of 3.5 slm. After the regrowth, the sample was subjected to channel waveguide formation. A SiO₂ mask was deposited using the PECVD technique (SAMCO, PD-10). The photoresist patterns were fabricated using a maskless lithography system (Nano System Solutions, DL-1000). Dry etching processes for GaN and SiO₂ were performed using ICP-RIE (ULVAC, NE750) and CCP-RIE (SAMCO, RIE-10N), respectively.

A nano three-dimensional optical interferometry topography system (Hitachi High-Tech, VS-1800) and atomic force microscopy (AFM; Hitachi High-Tech, S-image) were employed to observe surface morphology. The crystalline quality was characterized using an XRD system (Malvern Panalytical, X'pert Pro) with a Cu Kα₁ X-Ray radiation source

($\lambda = 1.54 \text{ \AA}$) and a Ge 220 double-crystal monochromator. The cross-sectional structure of the fabricated channel waveguide was observed using a field-emission SEM (Hitachi High-Tech, S-4300).

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

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

Keywords

GaN, metalorganic vapor-phase epitaxy, photon pair generation, polarity

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