Nanopatterned epitaxy of non-polar Ga$_{1-y}$In$_y$N layers with caps and voids

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Abstract

Low-defect density non-polar $a$-plane Ga$_{1-y}$In$_y$N layers on $r$-plane sapphire substrates are reported by implementing self-assembling nanopatterning in metalorganic vapor phase epitaxy. Nanopillar capping and void formation in the regrowth lead to a 90% defect reduction. An ex-situ Ni layer transforms into a nanoisland etch mask to pattern GaN templates. $a$-Plane GaN and Ga$_{1-y}$In$_y$N layers with an InN content in the range of $y = 0.04 – 0.11$ are then regrown. Both exhibit a low density of basal-plane stacking faults of $(4.6 \pm 1.3) \times 10^4$ cm$^{-1}$ by transmission electron microscopy analysis.

Growth parameters and template pattern are discussed by help of an X-ray rocking curve analysis. We find pattern fill factor and V/III ratio to dominate the defect reduction. Resulting layers should enable efficient long-wavelength light-emitting and solar cell devices.

Introduction

To extend performance of GaN-based light emitting diodes (LEDs) and solar cells from their 450 nm sweet spot of performance into the amber and red spectral region, it is important to improve the crystalline quality of Ga$_{1-x}$In$_x$N quantum well (QW) layers with higher InN content $x$. Typically such layers are grown on GaN, i.e. Ga$_{1-x}$In$_x$N/GaN, which leads to an increasing lattice mismatch with InN content, increased defect densities, and increasing piezoelectric polarization in c-plane growth. We
therefore developed templates of Ga$_1$yIn$_y$N that should replace the GaN layers, enabling Ga$_1$yIn$_y$N/Ga$_1$zIn$_z$N structures (where $x > y$) and thereby reduce all those detrimental aspects. Moreover, the templates use the non-polar $\alpha$-plane growth.

Conventional c-plane GaN-based heterostructures experience very high piezoelectric polarization due to lattice mismatch strain. Polarization fields of 1.2 MV/cm in c-plane Ga$_{0.84}$In$_{0.16}$N quantum wells (QWs) grown on GaN have been measured, and indirect estimates reach even higher values. Such fields spatially separate charge carriers within the QWs by the quantum confined Stark effect and thereby reduce radiative recombination. To compensate, QWs are kept to a narrow width below 3 nm. Such low recombination volume, however, leads to high excess carrier densities which in turn promotes non-radiative Auger recombination and decreased efficiency when emission wavelength is extended beyond the blue spectral region. Piezoelectric fields, however, can be eliminated by epitaxy on non-polar crystal orientations $\alpha$-plane or $m$-plane. Such non-polar QWs can then be increased in width, reducing carrier densities and their associated Auger losses. Corresponding structures also show vastly improved wavelength stability under variation of drive current density.

With higher In-content Ga$_1$yIn$_y$N QWs on GaN, and increasing lattice mismatch, resulting strain leads to relaxation and induces high densities of extended defects in non-polar just as in polar growth. Therefore, non-polar $\alpha$-plane GaN as commonly grown on $r$-plane sapphire usually exhibits a high density of basal-plane stacking faults (BSFs) in the range of 3.8 – 30 x 10$^5$ cm$^{-2}$, which so far limit its usefulness for high-efficiency light emitters.

To overcome both, piezoelectric polarization and the strain build-up in wide wells, we implement non-polar $\alpha$-plane growth in strain-relaxed Ga$_1$yIn$_y$N/GaN growth on $r$-plane sapphire. Strain relaxation can commonly be attained in regrowth on patterned templates. Such patterning can be applied to either a conventionally-grown GaN layer or to the substrate material itself. Typically highly symmetric structures are prepared in photolithographic or nanolithographic
processing steps that expose very specific crystallographic planes on the micrometer or nanometer length scale to limit regrowth to a single crystallographic plane. We, however, find that a self-assembled patterning into nanoislands is a cost-effective, easily implemented alternative of satisfactory performance. Using a self-assembled Ni nano etching mask we prepare α-plane GaN templates on r-plane sapphire. Transmission electron microscopy (TEM) analysis of α-plane GaN and Ga1−yInyN layers regrown on these nanopatterned templates reveals over 90% decrease in extended defect density relative to commonly reported values in planar growth of α-plane GaN on r-plane sapphire.

Experimental

Nanopatterning by self-assembled Ni nanoislands is used to produce templates for Ga1−yInyN regrowth (see schematic process flow in Figure 1). Metal-organic vapor phase epitaxy (MOVPE) on r-plane sapphire is initiated by growth of an AlN buffer layer followed by approximately 3 μm of α-plane GaN. Growth is accomplished using trimethylgallium (TMG) and ammonia (NH₃) as the group III and V precursors, respectively. To serve as an n-type device layer, the upper 600 nm of GaN is silane doped to a donor concentration of 1 x 10¹⁹ cm⁻³. On top, SiO₂ is deposited ex-situ by plasma-enhanced chemical vapor deposition (PECVD) to a thickness of 100 - 300 nm as measured by spectral reflectance. Next, a thin (10 - 30 nm) film of Ni is deposited by electron beam evaporation. The Ni thickness was monitored in situ by a quartz crystal microbalance. The samples are then subjected to rapid thermal annealing (RTA) in a N₂ environment at 850°C for one minute. This RTA process causes the Ni to agglomerate on the SiO₂ surface producing individual Ni nanoislands. These Ni nanoislands are then used as a hard mask during inductively coupled plasma reactive ion etching (ICP-RIE) through the SiO₂ and partially into the GaN. GaN etching proceeds until a depth in the range of 300 – 900 nm has been achieved. Thereafter, the Ni is removed using nitric acid. At this
point, the template surface consists of GaN pillars capped with SiO₂ across the entire substrate wafer surface.

These patterned templates are then reloaded into the reactor for epitaxial regrowth of GaN or Ga₁₋ₓInₓN. Regrowth of GaN was performed under a V/III ratio of 133, a temperature of 1040°C and a pressure of 10 kPa. All Ga₁₋ₓInₓN films were grown at temperatures of 753 – 820°C, pressures of 3.3 – 13 kPa, and flows of TMG of 41.8 – 83.6 μmol/min, TMI of 10.8 – 21.6 μmol/min and NH₃ of 4.46 – 313 mmol/min.

Figure 1: Schematic of Ni self-assembling nanopatterned GaN templates, here applied to non-polar α-plane growth of GaN and Ga₁₋ₓInₓN.

Scanning electron microscopy (SEM) is used to verify and quantify template pattern characteristics. Regrown GaN and Ga₁₋ₓInₓN layers are then analyzed by TEM and X-ray diffraction (XRD) using Cu-Kα radiation.

Results

Nanopatterned Template

Initial α-plane GaN films grown on r-plane sapphire are found to exhibit a (1120) rocking curve linewidth of 600 – 700 arcsec (FWHM). This compares very favorably to those reported in the literature: 600 – 2,000 arcsec,¹⁰ 731 – 1202 arcsec,¹¹ and 900 – 2,800 arcsec.²² Their surfaces are
then patterned using the Ni nanomask. By varying the Ni film thickness as deposited, island size and
density after RTA can be varied (Figure 2). For film thicknesses of 5 to 15 nm the island size was
shown to increase linearly as implemented here.

For the purpose of regrowth on the so prepared templates, we find the fill factor and lateral
size of the etched pillars as meaningful quantifiable metrics. We define them as the fraction of the
wafer surface covered by SiO$_2$-capped GaN pillars and the average surface area of the top faces,
respectively. Both values for several templates as a function of deposited Ni thickness are shown in
Figure 3. By this method, template fill factors ranging from 0.5 to 0.8 and average island sizes from
4,000 nm$^2$ to 100,000 nm$^2$ have been produced. Interestingly, 10 nm and 30 nm deposited Ni films
result in similar fill factors, but vastly different island sizes (Figure 3).

![Increasing Ni Thickness (nm)]

Figure 2: SEM top-view images of fully-fabricated nanopatterned templates as a function of deposited Ni film thickness. The fill factors (FF) are indicated.
Figure 3: (a) Average pillar size and (b) average template fill factor as a function of deposited Ni film thickness. Pillar size is found reasonably controlled by the thickness of the deposited Ni films.

α-Plane GaN on Nanopatterned GaN

As a starting point, GaN was regrown on top of nanopatterned α-plane GaN template on r-plane sapphire as described above. A preliminary assessment of the crystalline quality and the performance of resulting LED structures (mesa size 1 mm$^2$) have been provided in prior work.$^{20}$ There, the linewidth of XRD rocking curves showed a reduced anisotropy as a function of azimuth. This indicated an improved crystal quality of the regrown α-plane GaN when compared to the α-plane GaN without the patterning and regrowth steps. LED light output power was comparable to that of equivalent LED structures homoepitaxially grown on bulk α-plane GaN substrates.$^{20}$ Prior work had shown that LEDs in homoepitaxial GaN growth can far outperform conventional heteroepitaxial structures in terms of light output, whether operated electrically or pumped optically.$^{25}$ Thus, our LED results of nanopatterned heteroepitaxy performing at the level of homoepitaxially grown devices is most encouraging. Typically it is the reduction of extended defect densities that gives homoepitaxial growth on bulk GaN the performance advantage.$^{25}$ Therefore it is important to quantify the density of extended defects in these materials and elucidate the mechanisms of any defect reduction.
We use bright-field (multiple diffraction vectors) cross-sectional TEM to examine the GaN of the original, as well as the regrown layers (Figure 4). The region at the patterned interface shows a SiO₂ cap 200 nm thick and 200 nm wide. The interface of r-plane sapphire substrate and original GaN lies outside the visible frame towards the lower right. The material below the SiO₂ cap is interpreted as the elongated pillar of the original GaN. Its outer boundaries are sketched in white (Figure 4). The material surrounding the pillar and SiO₂ cap should therefore be the regrown GaN.

Figure 4: A bright-field cross-sectional TEM image of a GaN pillar (highlighted in white) with a SiO₂ cap surrounded by regrown GaN. The SiO₂ cap apparently blocks extended defects in the original GaN from propagating into the regrown material. The inset shows several diffractions contributing to the image.

From an XRD analysis (data not shown here, ²⁰) and zone-axis selected-area diffraction (SAD) patterns we find that regrown GaN (1120) α-planes lie parallel to the substrate surface ²⁰ and the direction of view is found to be [1100]. Hence, the growth direction is [1120] and lies along the long side of the original GaN pillar (see coordinates in Figure 4). Apparent dark lines oriented along the [1120] growth direction (Figure 4) correspond to extended defects threading with the layer growth. Tilting the specimen off the [1100] axis towards or away from [0001] produces a shadowing effect for most of the observed defects. This indicates that defects extend through the thickness of the specimen, are two-dimensional in nature, and the apparent lines are only the edges of the
faulted regions. These combined findings are well in agreement with those of other works in a-plane GaN\textsuperscript{10,14,26} assigning them to BSFs bound by partial dislocations. Apparently, the SiO\textsubscript{2} caps are very effective in blocking BSFs from propagating into the regrown GaN (Figure 4).

BSF densities in \(\alpha\)-plane GaN on \(\varphi\)-plane sapphire in the range of \(3.8 - 30 \times 10^5\) cm\(^{-1}\) are commonly reported for non-polar GaN films.\textsuperscript{14,15,16} For a representative quantification several areas across the regrown GaN were analyzed by TEM (Figure 5) and a stacking fault density of \((4.6 \pm 1.3) \times 10^4\) cm\(^{-1}\) was found. This represents a more than 90\% reduction in BSF density across the entire regrown GaN over the common literature values achieved by lateral epitaxial overgrowth averaging wing and window regions.\textsuperscript{27}

![Figure 5: Bright-field TEM cross-sectional images of various locations within the regrown GaN material showing consistently low BSF density throughout the specimen.](image)

BSFs are the prevalent extended defects in non-polar GaN.\textsuperscript{14} TEM analyses of \(\alpha\)-plane GaN grown on \(\varphi\)-plane sapphire reveal the presence of BSFs primarily at coalescence boundaries of separately-nucleated GaN islands.\textsuperscript{14,28} This is similar to the case of threading dislocations (TDs) in \(c\)-plane GaN grown on \(c\)-plane sapphire which are also primarily observed at coalescence boundaries.\textsuperscript{29,30,31} By the nature of the fault in the stacking sequence one distinguishes four types of BSFs – three intrinsic \(I_1\), \(I_2\) and \(I_3\) and one extrinsic type \(E\).\textsuperscript{16,32} Type \(I_3\) were found in experiment to
be the most dominant BSFs in non-polar GaN.\textsuperscript{16, 28, 33} This is compatible with density functional theory showing that I\textsubscript{1} BSFs have the lowest formation energy in both GaN and InN.\textsuperscript{32} BSFs are bounded by either prismatic stacking faults or, more likely, partial dislocations.\textsuperscript{14, 16, 28, 26} In non-polar GaN such partial dislocations have been shown to correspond to dark spots in cathodoluminescence images.\textsuperscript{28, 26} This suggests that they act as non-radiative recombination centers in non-polar GaN. Therefore, stacking faults are detrimental to device performance and we will analyze their occurrence and distribution in the following discussion.

\textit{\textbf{a-plane Ga\textsubscript{1-y}In\textsubscript{y}N on Nanopatterned GaN}}

Since the goal is a smaller bandgap for emitters and absorber, Ga\textsubscript{1-y}In\textsubscript{y}N layers were regrown on the nanopatterned GaN templates and analyzed. A characterization by photoluminescence (PL) at room temperature confirms the well-known dominance of the spectrum by a 30 – 50 nm peak attributed to the Ga\textsubscript{1-y}In\textsubscript{y}N layer (Figure 6(a)). Its peak wavelength in the range 400 – 500 nm is well controlled by variation of the Ga\textsubscript{1-y}In\textsubscript{y}N growth temperature in the range of 850 – 750 °C, respectively (Figure 6(b)). A weak high energy shoulder is subject of further study.

![Figure 6: PL characterization of a-Ga\textsubscript{1-y}In\textsubscript{y}N layers regrown on nanopatterned a-GaN. (a) PL spectra for various growth Ga\textsubscript{1-y}In\textsubscript{y}N temperatures. (b) Interpretation of peak emission wavelength versus Ga\textsubscript{1-y}In\textsubscript{y}N growth temperature.](image-url)
X-ray Diffraction Analysis

To estimate the InN fraction of the regrown Ga$_{1-y}$In$_y$N layers, XRD (1120) 2θ-ω data (not shown) was interpreted. From the 2θ peak value the interplanar spacing of the (1120) Ga$_{1-y}$In$_y$N planes, $d_{1120}$ was determined using Bragg’s law. It is related to the out-of-plane lattice constant, $a$, by

$$d_{hkl} = \left[ \frac{4(h^2 + k^2 + lh)}{3a^2} \right]^{1/2} + \left[ \frac{l^2}{c^2} \right]^{1/2},$$

where $d_{hkl}$ is the spacing of the planes with indices $h$, $k$, $l$ and $a$ and $c$ are the lattice constants of the wurtzite unit cell. Since in this case $l = 0$, the in-plane lattice constant, $c$, is not considered here. Due to the large layer thickness of at least a couple hundred nm, it is most reasonable to assume the majority of the Ga$_{1-y}$In$_y$N layers to be relaxed. We therefore reasonably assume no deformation along the in-plane [1100] m-direction. This is justified by the large thickness of the layer, well exceeding any reasonable assumption of the critical layer thickness. We then apply Vegard’s law assuming this $a$ lattice constant to vary linearly with composition from GaN to InN. In this way we find InN fractions of 0.04 to 0.11 across the set of samples. Under the unreasonable assumption of fully coherent strain, the unreasonably low values of 0.024 to 0.07 would be the result. Such strain however, could not be sustained throughout the thickness of the layers.

The presence of BSFs in α-plane GaN can be evaluated through PL studies where the extent of BSFs present can be correlated to the intensity of light emission at 3.42 eV. We find the PL spectra of our samples to be dominated by rather wide peaks as typical for the ternary Ga$_{1-y}$In$_y$N layers. An analysis in terms of contributions by BSFs in analogy to GaN therefore seems not possible at this point. Instead, to help judge the crystal quality of the regrown Ga$_{1-y}$In$_y$N layers we compare to the GaN of the underlying template by means of XRD. The method has been demonstrated for the case of m-plane GaN in the literature with excellent agreement to data obtained from TEM. In the case of α-plane GaN, it is known that for diffraction conditions of type $h0\bar{h}0$, both $h = 1$ and $h = 2$ are sensitive to $I_1$ and $I_2$ BSFs while the $h = 3$ condition is insensitive. This insensitivity of the 30̅30 diffraction is due to the fulfillment of the invisibility criterion $g_{hkl} \cdot R_F = n$, where $n = 0, \pm 1, \pm 2, \ldots$.
and \( \mathbf{g}_{hkl} \) is the reciprocal lattice vector of the planes with indices \( h, k, l \). \( \mathbf{R}_f \) is a vector representing a translation of the crystal lattice due to the fault in the stacking sequence.\(^{16}\) XRD reciprocal space maps (RSM) of the regrown Ga\(_{1-y}\)In\(_y\)N layers under 20\(2\) and 30\(3\) diffraction conditions are shown in Figure 7. The distance from the origin \( (Q_x = Q_y = 0) \) in reciprocal space to a peak corresponding to planes with spacing \( d \) is given by \( \frac{\lambda}{2d} \) where \( \lambda \) is the X-ray wavelength. The vertical separation of Ga\(_{1-y}\)In\(_y\)N and GaN peaks in the RSMs should increase with the order of diffraction. For better clarity we therefore analyze the 20\(2\) rather than the 10\(1\)0 and compare to the 30\(3\)0 diffraction.

Figure 7: 30\(3\)0 and 20\(2\)0 XRD RSMs of Ga\(_{1-y}\)In\(_y\)N regrown on a patterned GaN template. Peaks from both, the regrown Ga\(_{1-y}\)In\(_y\)N and underlying GaN template are present in each RSM. BSFs contribute in horizontal streaking of the 20\(2\)0 peaks, but are absent in the 30\(3\)0 peaks. Scales of \( Q_x \) and \( Q_y \) axes are identical across both RSMs for a direct comparison of angles and peak widths.
The RSMs include peaks from both, the regrown Ga$_{1-x}$In$_x$N and the underlying GaN template, as indicated (Figure 7). Both 2020 diffraction peaks are found horizontally stretched in comparison to the 3030 peaks. From the invisibility criterion, we attribute this stretching to the presence of BSFs. Therefore, comparing ratios of the widths of 2020 diffraction peaks to the width of the 3030 peaks gives a measure of stacking fault densities. The same mechanism, however, also applies to XRD rocking curve linewidths if measured along that width. 2020 and 3030 rocking curve linewidths from all Ga$_{1-x}$In$_x$N samples are plotted against their respective InN contents in Figure 8. We observe a positive correlation between Ga$_{1-x}$In$_x$N rocking curve linewidth and InN content. At this point it is not apparent if this is caused by increasing InN content, a degradation in crystal quality, or both.

![Graph showing 2020 and 3030 rocking curve FWHM values as a function of InN content for the regrown Ga$_{1-x}$In$_x$N layers. Lines of best fit are shown along with their coefficient of determination ($R^2$) values.](image)

Factors relevant for the XRD linewidth are layer thickness\textsuperscript{38} and, specifically for non-polar GaN, density of extended defects, surface roughness, wafer curvature, and mosaic in-plane twist and out-of-plane tilt.\textsuperscript{39} When mosaic twist and tilt dominate, a Williamson-Hall analysis can be
performed\textsuperscript{11,16} leading to a quantitative measure of the BSF density.\textsuperscript{16} Other work, however, has shown that such an analysis can be off, reporting only half the density seen in TEM.\textsuperscript{10} In our work, the regrown Ga\textsubscript{1-x}In\textsubscript{x}N material is roughly an order of magnitude thinner than the GaN template, disallowing their quantitative comparison by Williamson-Hall. This is well apparent in the much stronger and narrower line width of the GaN template over that of the regrown Ga\textsubscript{1-x}In\textsubscript{x}N material (Figure 7). We therefore resort to a simpler qualitative interpretation as follows.

For each material, 20\textsuperscript{2} and 30\textsuperscript{3} rocking curve data were obtained from both the GaN and Ga\textsubscript{1-x}In\textsubscript{x}N diffraction peaks. Such rocking curves well approximate horizontal scans in the RSMs of Figure 7. Since the diffraction planes are not parallel to the material’s surface, we use the skew-symmetric rocking curve geometry with the incident X-ray beam projected along the [0001] axis of the GaN template.\textsuperscript{11,46} The resulting peaks were fitted with Gaussian functions, from which full width at half maximum (FWHM) values were extracted. The ratios of the 20\textsuperscript{2} linewidths to the respective 30\textsuperscript{3} linewidths then provide a measure for comparison of the density of BSFs present in the regrown Ga\textsubscript{1-x}In\textsubscript{x}N and, separately, the underlying GaN template. This measure is qualitative only since a quantitative correlation to BSF density is not known. An inherent assumption is that BSFs are the only broadening factor differentiating the two peak widths. While those mentioned factors are known to broaden the peak widths of XRD rocking curves, none of those factors are known to preferentially broaden the 20\textsuperscript{2} diffraction peak over the 30\textsuperscript{3} peak (or vice versa), with the exception of I\textsubscript{1} and I\textsubscript{2} BSFs due to the invisibility criterion. The effect of these non-preferential linewidth broadening factors are assumed to be nullified when the ratio is taken between the 20\textsuperscript{2} and 30\textsuperscript{3} rocking curve peak widths. Therefore, in this comparison a lower 20\textsuperscript{2} FWHM / 30\textsuperscript{3} FWHM ratio indicates a lower density of I\textsubscript{1} and I\textsubscript{2} BSFs.
X-ray Diffraction Basal Stacking Fault Density Comparison

The ratios of the rocking curve widths (202 FWHM / 3030 FWHM) for several Ga$_{1-y}$In$_y$N samples regrown under a range of relevant growth parameters are summarized in Figure 9. The data is sorted along the V/III ratios of the regrown Ga$_{1-y}$In$_y$N (Figure 9 (a)). A subset of the same data is sorted along the template fill factor (Figure 9 (b)) and the full dataset is plotted against interpreted InN fraction of the regrown Ga$_{1-y}$In$_y$N layer (Figure 9 (c)).

We find that values for the underlying GaN remain rather constant throughout the data set in either of the three sorting orders. This is expected since V/III ratios were kept constant for all GaN templates and only varied for the Ga$_{1-y}$In$_y$N regrown portions. In comparison, all values for the regrown Ga$_{1-y}$In$_y$N layers were below those of their respective GaN template values. Moreover they show a trend towards lower values with increasing V/III ratio (Figure 9 (a)), with increasing fill factor (Figure 9 (b)), and – to a much lesser extent – with increasing interpreted InN fraction but at a much lower level of correlation ($R^2 = 0.0446$) (Figure 9 (c)). A number of Ga$_{1-y}$In$_y$N samples were regrown at a V/III ratio of 212 and therefore lead to some data clustering in Figure 9 (a). The spread of this clustering is well accounted for by plotting that subset against template fill factor (Figure 9 (b)).

![Figure 9: 202 FWHM / 3030 FWHM ratio of both GaN templates and regrown Ga$_{1-y}$In$_y$N materials as a function of (a) V/III ratio during Ga$_{1-y}$In$_y$N regrowth, (b) template fill factor for the case where V/III = 212, and (c) InN content of the regrown Ga$_{1-y}$In$_y$N. The solid lines shown the best linear fits. The fit qualities are indicated by the coefficient of determination $R^2$, where a value 1 indicates perfect correlation and 0 indicates no correlation.](image-url)
From the preceding we identify V/III ratio and template fill factor as the relevant control parameters for Ga$_{1-y}$In$_y$N regrowth in terms of linewidth ratio reduction while InN fraction in itself is not. We therefore reanalyze the linewidth ratios versus V/III ratio graph of Figure 9 (a) by template fill factor in Figure 10 (a). A lower linewidth ratio is found for both, increasing V/III ratio and increasing template fill factor, in agreement with the original trends (Figure 9).

The role of growth temperature is revealed in Figure 10 (b). Template FF = 0.73 was regrown at V/III = 212 under two different temperatures. The material of the lower temperature exhibits the lower linewidth ratio. Similarly, the role of growth pressure is revealed in Figure 10(c). Template FF = 0.65 was regrown at V/III = 212 under two different growth pressures. Here the material of the higher pressure shows the lower linewidth ratio. Accordingly, besides a high V/III ratio and high template fill factor, a lower growth temperature and higher growth pressure are desirable for a low linewidth ratio and low defect density.

![Figure 10: 2020 FWHM / 3050 FWHM ratio of the GaN templates and Ga$_{1-y}$In$_y$N regrowth as a function of (a) V/III ratio during Ga$_{1-y}$In$_y$N growth grouped by template fill factor (FF), (b) Ga$_{1-y}$In$_y$N growth temperature for the case where V/III = 212 and FF = 0.73, and (c) Ga$_{1-y}$In$_y$N growth pressure for the case where V/III = 212 and FF = 0.65. Templates correspond to those in Figure 2 identified by their fill factors.]

For the Ga$_{1-y}$In$_y$N regrown on templates with FF = 0.73 and FF = 0.75, there is a large difference in linewidth ratios (Figure 10(a)), even beyond what might be expected based on differences in V/III ratio. However, by choice of the original Ni layer thickness, island size of template FF = 0.75 is twenty times larger than that of template FF = 0.73 (Figure 3). This suggests
that the larger pillar of size $1.3 \times 10^5$ nm$^2$ (400 nm diameter) may benefit the reduction of extended defects when compared to the smaller pillars of size $6.5 \times 10^3$ nm$^2$ (90 nm diameter). The effect, however, is subordinate to that of a high V/III ratio and high template fill factor.

Transmission Electron Microscopy Analysis

The preceding XRD analysis indicated that fill factor and V/III ratio during Ga$_{1-y}$In$_y$N regrowth strongly affect defect reduction on the patterned template. Therefore, the materials with the highest and lowest V/III ratio during Ga$_{1-y}$In$_y$N growth are further investigated using TEM. Bright field cross-sectional TEM images of these two samples are shown in Figure 11 as viewed along the [1100] direction. Identifiable features are the GaN pillars, SiO$_2$ caps, voids, and the regrown Ga$_{1-y}$In$_y$N material, as indicated (Figure 11(a), (b)). Extended linear defects can be identified in both the GaN template portion and the regrown Ga$_{1-y}$In$_y$N. In addition, the regrown Ga$_{1-y}$In$_y$N layer shows a variegated contrast. This is commonly observed in Ga$_{1-y}$In$_y$N alloy films in the literature and interpreted as a direct consequence of the various configurations of the In atom within the alloy. Whether such variations here reach beyond the binomial distribution of a uniform alloy, e.g. driven by spinodal decomposition, cannot be concluded from this data.41

The surface of the Ga$_{1-y}$In$_y$N films exhibit a faceted appearance in SEM images (not shown here). The TEM data of the material grown at V/III = 5900 (Figure 10 (b)) includes the as-grown surface which allows a crystallographic analysis of the faceted appearance. The facet angles were determined in reference to the top GaN surface of the template underneath the SiO$_2$ cap known to be $\alpha$-plane (Figure 11(b)). We find members of the \{11\overline{2}2\}, \{20\overline{2}1\}, \{1\overline{0}\overline{1}2\} and \{10\overline{1}1\} families of planes as likely candidates in agreement with those commonly seen in the literature.10, 12, 13, 22, 42, 43, 44

The vertical faces of the template pillars present a large variety of potential growth planes. However, growth studies in the literature have shown that only a small subset of growth planes are kinetically favorable during GaN growth prior to coalescence, particularly the (000\overline{1}) and \{10\overline{1}1\}
We expect these same crystal facets to dominate the lateral growth from the template pillar sidewalls prior to coalescence. Evidence of \{10\bar{1} 1\} planes is observed here (Figure 11(b)) possibly remaining from growth which occurred prior to film coalescence.

*Figure 11: Cross-sectional TEM of a-plane Ga$_{1-y}$In$_y$N materials regrown at (a) V/III = 70 and (b) V/III = 5900 on nanopatterned GaN-on-sapphire templates. The Ga$_{1-y}$In$_y$N surface is visible in (b) and exhibits a faceted appearance with planes as indicated.*

Lines of dark contrast oriented along the [11\bar{2}0] growth direction (Figure 11 (a) and (b)) are interpreted as BSFs. Their densities were determined for each, the underlying GaN and the regrown Ga$_{1-y}$In$_y$N. For the GaN templates we find $2.0 \times 10^5$ cm$^{-1}$ (material with regrowth at V/III = 70) and $2.4 \times 10^6$ cm$^{-1}$ (material with regrowth at V/III = 5900). These densities are in good agreement with
literature data for such α-plane GaN grown on τ-plane sapphire.14,15,16 The BSF densities of the regrown Ga1−yInyN materials, however, are found to be significantly lower at 7.8 \times 10^4 \, \text{cm}^{-1} and 5.0 \times 10^4 \, \text{cm}^{-1} for V/III ratios of 70 and 5900, respectively. The Ga1−yInyN BSF density of the higher V/III ratio material is in good agreement with that of the regrown GaN (Figure 4). Moreover, it is significantly less than the BSF density of the low V/III material in spite of the similar densities in their respective templates. These results suggest that growth at a high V/III ratio is beneficial in terms of BSF reduction in the regrown materials, in agreement with the XRD analysis.

Cross-sectional TEM images zooming to the nanopatterned regions of these materials are shown in Figure 12 (a) (V/III = 70, FF = 0.73) and Figure 12(b) (V/III = 5900, FF = 0.75). They show very similar size and spacing of the SiO2 caps, which is consistent with the very similar template fill factors. On the other hand, in addition to the much lower density of the extended defects as discussed above, we here observe a larger number and a larger size of voids for the material grown at the higher V/III ratio. In fact, we hypothesize that the higher V/III ratio during Ga1−yInyN regrowth enhances the formation of voids and thereby reduces the density of extended defects.
Figure 12: Cross-sectional TEM of Ga$_{1-y}$In$_y$N regrown on nanopatterned GaN templates under V/III ratios as indicated. The voids of the material regrown at a V/III ratio of 5900 ((b)) are larger and more numerous than those regrown at V/III = 70 ((a)).

Discussion

Two metrics were found useful to quantify the templates’ pattern characteristics: fill factor and average pillar size. There is a clear correlation between deposited Ni thickness and average pillar size (Figure 3 (a)). The formation of Ni islands occurs by self-assembly, and fill factor and average pillar size were found highly reproducible.

We find the rocking curve linewidth ratios of every regrown Ga$_{1-y}$In$_y$N material is below that of the original GaN template. This is evidence of defect reduction in the regrown Ga$_{1-y}$In$_y$N relative to the GaN templates. We found only a weak correlation between defect reduction and InN
fraction. Consequently defect reduction must be attributed to the use of the nanopatterned templates and the approach may prove suitable for even higher InN fraction alloys.

The X-ray analysis of the regrown Ga$_{1-y}$In$_y$N reveals that defect reduction increases with fill factor, the measure of how much of the template surface is covered with SiO$_2$ caps. These features are therefore found to be effective in preventing the propagation of BSFs into the regrown GaN and Ga$_{1-y}$In$_y$N as evidenced in TEM. This mechanism alone cannot explain the large magnitude of defect reduction. However, the SiO$_2$ caps are found to occur in a staggered fashion with voids formed at the bottom of the template pillars.$^{21,45}$ TEM reveals these staggered features result in a very uniform decrease in the BSF density of the regrown GaN (4.6 x 10$^4$ cm$^{-1}$) and regrown Ga$_{1-y}$In$_y$N (5.0 x 10$^4$ cm$^{-1}$) by 75% compared to our GaN templates (2.0 x 10$^5$ cm$^{-1}$) and over 90% when compared to commonly reported BSF densities of α-plane GaN grown on r-plane sapphire (3.8 – 30 x 10$^5$ cm$^{-1}$).$^{14,15,16}$ The uniformity of BSF reduction is an important advantage over conventional lateral epitaxial overgrowth where defects cannot be stopped in the window region.$^{17,27}$

The X-ray analysis also finds that Ga$_{1-y}$In$_y$N regrown at a higher V/III ratio tends to show greater defect reduction than regrowth at a lower V/III ratio. It is known that a high V/III ratio promotes growth along the vertical [1120] direction in α-plane GaN.$^{46}$ Furthermore, our data shows that defect reduction is enhanced with decreasing growth temperature and increasing growth pressure (Figure 10 (b) and (c), respectively) which are also known to enhance vertical growth in α-plane GaN.$^{46}$

Ga$_{1-y}$In$_y$N growth from the GaN pillar sidewalls with an enhanced vertical component could lead to merging growth fronts which would quickly cut off the supply of precursors to the volume near the bottom of the pillars resulting in closed-off voids, found here to effectively block extended defects of the template from continuing into the regrown material. This can explain the observed trend of defect reduction with increasing V/III ratio during regrowth of Ga$_{1-y}$In$_y$N.
The comparison of Ga$_{1-y}$In$_y$N defect reduction between materials regrown on templates of similar fill factor (FF = 0.75 and FF = 0.73, Figure 10 (a)), but different pillar size (Figure 2) suggests that larger pillars are more beneficial to defect reduction than smaller pillars. When regrown Ga$_{1-y}$In$_y$N growth fronts from neighboring pillars meet each other, there is the potential for mismatch between them which could lead to the formation of new BSFs in the regrown material. Thus, the more merging growth fronts, the more defects are expected in the regrown material. The number of merging growth fronts in the regrown material would be expected to be much higher for templates consisting of many small pillars than for templates consisting of fewer large pillars. This can explain the enhanced defect reduction of the material grown on the templates with larger average pillar size for a given fill factor. Increasing template fill factor from FF = 0.52 to FF = 0.75 with V/III ratio in the range of 850 – 2500 results in a larger decrease in GaInN linewidth ratios than both increasing V/III from 210 – 2500 at FF = 0.52 and from 850 – 5900 at FF = 0.75 (Figure 10 (a)).

Accordingly we find fill factor as the primary factor to reduce defect density, then V/III ratio as a secondary factor, while growth temperature (Figure 10 (b)), growth pressure (Figure 10 (c)) and pillar size range lower in priority.

Conclusions

Nanopatterned $\alpha$-plane GaN templates were used to regrow $\alpha$-plane GaN and Ga$_{1-y}$In$_y$N. TEM analysis reveals the effectiveness of capping the template nanopillars with SiO$_2$ and the importance of void formation at the bottom of the pillars. Both are found to block the propagation of extended defects into the regrown material and together they cover the entirety of the template surface. Defect reduction in the regrown Ga$_{1-y}$In$_y$N increases primarily with increasing template fill factor. Defect reduction also increases with increasing V/III ratio and, to a lesser extent, increasing growth pressure and temperature, all of which are attributed to the formation of voids between template
pillars. Large SiO$_2$-capped GaN pillar size, obtained by choice of deposited Ni film thickness, helps to maintain the low defect density obtained via nanopatterning as merging growth fronts coalesce.

Here InN content of the materials does not appear to be relevant to the amount of defect reduction suggesting this growth method is a promising means to achieve even higher InN content Ga$_{1-x}$In$_x$N materials. This growth method demonstrates an effective and practical method for achieving high quality Ga$_{1-x}$In$_x$N which can be used as templates for the growth of efficient long-wavelength light-emitting structures and devices.

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(a) Wavelength (nm) vs. PL Intensity (Counts)

$\lambda_{\text{Peak}}$ (nm) $T = 300\text{K}$

$T_{\text{Growth}}$ ($^\circ\text{C}$)

(b) Growth Temperature ($^\circ\text{C}$) vs. Wavelength (nm)

a-GaInN/GaN