2010

Growth and characterization of non-polar GaN materials and investigation of efficiency droop in InGaN light emitting diodes

Xianfeng Ni
Virginia Commonwealth University

Follow this and additional works at: https://scholarscompass.vcu.edu/etd
Part of the Electrical and Computer Engineering Commons

© The Author

Downloaded from https://scholarscompass.vcu.edu/etd/2235

This Dissertation is brought to you for free and open access by the Graduate School at VCU Scholars Compass. It has been accepted for inclusion in Theses and Dissertations by an authorized administrator of VCU Scholars Compass. For more information, please contact libcompass@vcu.edu.
Growth and characterization of non-polar GaN materials and investigation of efficiency droop in InGaN light emitting diodes

Xianfeng Ni

A research dissertation submitted in partial satisfaction of the requirements for the degree of

Doctor of Philosophy

In
Electrical Engineering

Committee in charge

Prof. Hadis Morkoç (thesis advisor)
Prof. Alison Baski
Prof. Arvydas Matulionis (Lithuania)
Prof. Mikhail Reschikov
Prof. Supriyo Bandyopadhyay
Prof. Ümit Özgür

August 2010
# Table of contents

Table of contents ............................................................................................................................. ii  
Acknowledgement .......................................................................................................................... v  
List of tables ................................................................................................................................... vi  
List of figures ................................................................................................................................ vii  
Abstract ........................................................................................................................................ xvi  
Chapter 1. Introduction ................................................................................................................... 1  
  1.1 InGaN light emitting diode (LED) structure .......................................................................... 2  
  1.2 Carrier recombination in LEDs .............................................................................................. 3  
  1.3 The motivation of non-polar nitrides for light emitting applications .................................... 4  
  1.4 Materials growth procedures – metalorganic chemical vapor deposition (MOCVD) ........... 6  
  1.5 Epitaxial lateral overgrowth (ELO) ....................................................................................... 7  
  1.6 Methods for measurement of LED internal quantum efficiency (IQE) ............................... 10  
  1.7 Organization of this thesis ................................................................................................... 13  
Chapter 2. Epitaxial growth of non-polar GaN materials ............................................................. 15  
  2.1 Epitaxial growth of a-plane GaN on r-plane sapphire ......................................................... 15  
  2.2 Epitaxial lateral overgrowth of a-plane GaN on r-plane sapphire ....................................... 22  
  2.2a Experimental procedures .............................................................................................. 23  
  2.2b Scanning electron microscopy (SEM) images .................................................................. 24  
  2.2c X-ray Diffraction (XRD) results ....................................................................................... 27  
  2.2d Transmission electron microscopy (TEM) results ....................................................... 29  
  2.2e Atomic force microscopy (AFM) images .................................................................... 31  
  2.2f Near-field scanning optical microscopy (NSOM) ........................................................ 36  
  2.2g Photoluminescence (PL) results ................................................................................... 37  
  2.2h Conclusions .................................................................................................................. 40  
  2.3 Epitaxial lateral overgrowth of (1 100) m-plane GaN on m-plane 6H-SiC ......................... 41  
  2.3a Experimental procedures .............................................................................................. 41  
  2.3b Results and discussion ................................................................................................. 42  
  2.3c Conclusions .................................................................................................................. 48
5.1 Motivation .......................................................................................................................... 123
5.2 Experimental procedure ..................................................................................................... 124
5.3 Results and discussion ....................................................................................................... 125
5.4 Conclusion ......................................................................................................................... 130
Chapter 6. Future work ........................................................................................................... 132
  6.1 Preparation of large-area high-quality m-plane substrates ............................................. 132
  6.2 Hot-electron overflow model ............................................................................................ 133
  6.3 Enhancement of hole injection into active region ............................................................ 134
Appendix A: IQE from power dependent photoluminescence measurement .................... 136
References ................................................................................................................................... 141
Vita .............................................................................................................................................. 153
Acknowledgement

It is an extraordinary experience to have worked in VCU for my PhD study. My first and deepest appreciation must go to my thesis advisor Professor Hadis Morkoç. He offered me such a great chance to work on nitride materials and light emitting diodes, which has become one of the most exciting fields among all of the scientific and engineering topics. His deep thoughts and inspirational suggestions have guided me through the scientific discovering process of my PhD study, and will surely become my invaluable possessions which will continue to influence my future research. I am also grateful to Professor Ümit Özgür for his consistent and valuable effort to help me, to encourage me during my research, and also his valuable suggestions for issue solving.

I would like to thank my committee members: Prof. Supriyo Bandyopadhyay, Prof. Alison Baski, Prof. Mikhail Reshchikov, Prof. Arvydas Matulionis for their time, effort and comments. Especially I am indebted to Prof. Arvydas Matulionis for his great suggestions with the development of hot electron model of this thesis. It was a great pleasure and memory to have worked with him during his visit to VCU.

I would like to acknowledge Dr. Vitaliy Avrutin for his help and guidance for my research and equipment troubleshooting. I am also thankful to Dr. Jaesoong Lee for his help with the MOCVD system maintenance and valuable discussions. I also benefited from discussions with Drs. Y. Moon, R. Shimada, J. Xie, Y. Fu, X. Gu, Q. Fan and J. Leach. It is great pleasure to have worked with my lab mates: M. Wu, X. Li, E. Rowe, H. Liu, Amy Liu, S. Liu, F. Zhang, C. Zhu. Last, but not least, I would like to thank my family for their understanding and support.
List of tables

**Table 1.** FWHM values (in arcsec) of XRD rocking curves for (1122) GaN templates, ELO samples with mask stripes along the $a$-axis and $c$-axis of sapphire, and an $a$-plane GaN ELO sample for different rocking directions. ........................................................................................ 61

**Table 2.** Calculated electron overflow percentiles for a one-layer SEI, with varying SEI step height ($\Delta E_c$) and SEI thickness ($d$). ................................................................. 107
List of figures

Figure 1. Schematic for a typical InGaN LED structure in this thesis with a simplified conduction band edge profile shown on the right side. ................................................................. 3

Figure 2. Calculated band profiles in (5nm GaN)/(10nm Al0.1Ga0.9N) quantum wells by self-consistent effective mass Schrödinger-Poisson calculations. a, The very large electrostatic fields in the [0001] orientation result in a quantum confined Stark effect and poor electron-hole overlap. b, The [1 ̅00] orientation is free of electrostatic fields, thus true flat-band conditions are established................................................................. 5

Figure 3. Schematic representation of m-, a-, r- and c-plane of GaN. ........................................... 6

Figure 4. A schematic for MO source bubbler used in MOCVD system for providing MO source. The MO source mole flow rate is determined by (1) MO source temperature, (2) carrier gas flow rate, and (3) bubbler pressure (controlled by an upstream pressure controller). ............................ 7

Figure 5. Schematic for an ELO process: (a) stripe-shaped mask (typically SiO₂ or SiNₓ dielectric films) on planar GaN template was obtained through photolithography procedures with the open area (“window”) exposed for the regrowth process. (b) During regrowth step, the GaN material starts growth at the window regions, and expands both vertically and laterally. In contrast to the window regions, the overgrowth GaN above the mask regions which is normally called “wings” has very few defects. (c) After certain amount of time, the GaN wings will coalesce and form continuous films. A few defects will be generated at the coalescence boundary, where the two opposite wings coalesce. (d) The directions for the patterning of ELO mask for c-plane, a-plane and m-plane GaN ELO. It should be noted that in a-plane and m-plane GaN ELO, the Ga-polar wings have higher lateral growth rate than that of N-polar wings. So the coalescence boundary is not in the middle of the masked regions. ........................................... 9

Figure 6. SEM images of 0.9μm (1120) a-plane GaN grown on (1 ̅102) r-plane sapphire at 980°C, with 7000ccm NH₃, under (a) 30torr, (b) 200torr, respectively................................................................. 16

Figure 7. SEM images of (11₂₀) a-plane GaN grown under 30torr, at 1070°C, with NH₃ flow rates of (a) 7060ccm, (b) 4500ccm, (c) 1000ccm, respectively................................................................. 17
Figure 8. SEM images of (1120) a-plane GaN grown under 30torr with NH3 flow rate of 1000 sccm, with growth temperature being (a) 1050°C, (b) 1070°C, (c) 1090°C, respectively. ........... 18

Figure 9. Surface morphological evolution during early stage of a-plane GaN growth: (a) LT-GaN buffer, (b) 20nm GaN, (c) 50nm GaN, (d) 100nm GaN, (e) 300nm GaN, (f) 1.5μm fully coalesced GaN. Note that c direction is the stripe direction shown in (f), and all samples here have their c directions parallel to each other. ........................................................................................................ 20

Figure 10. XRD spectra using 2θ-ω scan for a-plane GaN grown with LT-AlN buffer layer on (1 102) r-plane sapphire................................................................................................................ 21

Figure 11. Surface morphology measurements of (1120) a-plane GaN films grown with HT-AlN buffer, by (a) AFM and (b) SEM measurements. ........................................................................................................ 21

Figure 12. Plan-view SEM images for sample A after (a) 0.5 h and (c) 3.0 h of growth. (b) Cross-sectional SEM image for sample A after 3 h of growth. (d) Cross-sectional SEM image for sample B after 2 h of growth at 1000 ºC. (e) Cross-sectional and (f) plan-view SEM images for sample B after a total of 5 h of growth. ........................................................................................ 26

Figure 13. Schematics for a-plane GaN ELO showing the origin of the height difference between two neighboring wings (not in exact proportion), with Ga- to N- polar wing width ratios of (a) 5:1 and (b) 1.6:1. The larger this ratio, the larger the height difference is between the two opposite wings at the coalescence fronts. (c) Schematic showing the inclination of the growth planes due to the 1.05° miscut of the r-plane sapphire towards its [0001] c-axis. The miscut of the substrate, and therefore, the resultant inclination of the GaN growth planes is the origin of the elevated meeting fronts observed from AFM. .............................................................................. 27

Figure 14. XRD rocking curve data for (a) sample A and (b) sample B with different φ angles. The dashed lines correspond to the multiple Gaussian fits to the rocking curve data. ............... 29

Figure 15. (2110) Kikuchi lines from large angle convergent beam electron diffraction at the meeting front of the two opposing wings for sample A. Note that the Kikuchi lines are shifted across the meeting front. ................................................................................................................. 30

Figure 16. Cross-sectional TEM images showing the window and wing regions for (a) sample A and (b) sample B. .................................................................................................................. 31
Figure 17. (a) 30 µm × 30 µm AFM image for sample B. (b) 4 µm × 4 µm AFM image near the window and N-polar wing boundary of sample B, showing different surface pit densities for the window and the wing. ................................................................................................................................. 34

Figure 18. (a) 20 µm × 20 µm AFM image for sample B after etching in a mixed H₃PO₄/H₂SO₄ (1:1) solution for 20 min at 200 ºC. (b) Cross-sectional TEM image for sample B showing the distributions of basal stacking faults in the wings, with most of them located near window regions. ........................................................................................................................................ 35

Figure 19. (a) AFM and (b) NSOM scans from a 40 µm × 40 µm area of a-GaN ELO sample B. (c) AFM and (d) NSOM scans from a 75 µm × 75 µm area of a-GaN ELO sample C (20 µm-stripe sample). The vertical scale bars in (a) and (b) correspond to 85 nm and 150 nm, respectively. [Ref. ] ....................................................................................................................................... 37

Figure 20. (a) PL spectra for sample B at 15 K and 300 K and for the control a-plane GaN sample at 15 K. (b) PL spectra for sample B at temperatures 15, 25, 50, 75, 100, 150, 200, 250, and 300 K (from top to bottom and vertically shifted for clarity). ........................................................................................................................................................................ 39

Figure 21. Normalized room temperature time-resolved PL for Sample B and a standard a-plane GaN template. Solid line represents the biexponential fit to data. The inset shows the near bandedge PL at room temperature and the effective PL decay times (τ_eff) at different photon energies. ........................................................................................................................................ 40

Figure 22. Off-axis XRD scans to determine the epitaxial relationships between m-plane GaN and m-plane 6H-SiC. ........................................................................................................................................ 43

Figure 23. (a) A typical SEM image of an m-plane GaN template grown on an m-plane SiC substrate. The directions were determined by XRD, (b) A tapping-mode AFM image (5 x 5 µm²) of an m-plane GaN template, with a RMS roughness of 2.8 nm. .................................................................................................................. 43

Figure 24. Plan-view SEM images for ELO sample with (a) 1h regrowth, (b) 2.5h regrowth, (c) 4.5h regrowth, respectively. (d) The schematic for the m-plane GaN ELO process. The inset in (c) shows a typical trench found in some areas of the sample. ................................................................................................. 45

Figure 25. A tapping-mode AFM image (4 x 4 µm²) of m-plane GaN ELO from the overgrown area (ΔZ=10 nm). ........................................................................................................................................ 46

Figure 26. PL spectra for an m-plane GaN ELO sample at 10K and 300K, with the inset showing the details of the near-band edge emission. ................................................................................................. 47
Figure 27. Schematic depiction of selective area $m$-plane GaN growth on patterned Si(112) substrates. Since growth is initiated on the vertical Si($\overline{1}1\overline{1}$) sidewalls, the threading dislocations propagate along the c-axis and not toward the surface. ................................................................. 50

Figure 28. An angled cross-sectional SEM image of a patterned Si(112) substrate ready for GaN epitaxy, where the vertical sidewall is covered by AlN. The top Si(112) plane and the tilted Si(111) plane are both masked with SiO$_2$. ........................................................................................................... 51

Figure 29. (a) Plan-view and (b) angled cross-sectional SEM images of an $m$-GaN sample grown on a patterned Si(112) substrate after a 3 h growth. (c) Enlarged cross-sectional SEM image showing the growth of the $m$-GaN initiated at the vertical Si($\overline{1}1\overline{1}$) sidewalls. ......................... 52

Figure 30. Tapping-mode AFM image ($\Delta z = 10$ nm) of an $m$-plane GaN sample (~12µm thick) grown on a patterned Si substrate. The AFM image indicates a very smooth surface (RMS roughness of ~0.3 nm over an area of 2 µm×2 µm) with clear atomic steps.................................................. 52

Figure 31. (a). On-axis XRD $\omega$-2$\theta$ scan showing that the GaN has (1$\overline{1}00$) $m$-plane orientation. The Si(112) plane does not have a diffraction peak due to diffraction extinction. The inset shows the off-axis scan results, suggesting that GaN(1$\overline{1}01$) is oriented 180º away from Si(111). (b) XRD rocking curves of GaN(1$\overline{1}00$) $m$-plane after a 3 h growth, rocking toward the GaN $a$-axis and $c$ axis....................................................................................................................................... 54

Figure 32. Room-temperature PL spectra measured with a He-Cd laser at different excitation densities for 6 nm-thick InGaN double heterostructure LED active layers on (a) $c$-GaN on sapphire and (b) $m$-GaN on Si. No blueshift is observed in the emission peak of the $m$-plane sample. The excitation power densities in (a) and (b) were 0.05, 0.15, 0.52, 1.0, 2.0, 2.5, 5.1 kW/cm$^2$. Figure (c) shows the internal quantum efficiencies of both samples extracted from the excitation dependence of the PL intensity using a Ti-sapphire laser (370nm) .............................................. 56

Figure 33. (a) SEM image of the $m$-plane InGaN sample on Si(112), (b) Spatial distribution of integrated CL intensity with wavelength from 362nm to 364 nm, which is attributed to BSF-related emission, (c) Spatial distribution of integrated CL intensity with wavelength from 384nm to 396nm, which is attributed to InGaN emission. Brighter regions in (b) and (c) indicate higher intensity. The scale bar for images (a), (b) and (c) is shown in (a). (d) Spatially integrated Spatially integrated CL spectrum from the image shown in (a). The results suggest that BSFs are
only located in the area where the vertical Si (111) sidewalls and the sample surface are intercepted.

Figure 34. Bright-field TEM image of a Nitrogen-polar wing GaN grown on patterned Si substrate.

Figure 35. (a) XRD 2θ-ω scan for GaN grown on m-plane sapphire, with the inset showing the off-axis ϕ scans with different ψ tilt angles (i.e., pole figure) for GaN (0002) and sapphire (01 1̅2) of GaN(1122) on m-plane sapphire. (b) A schematic depicting the epitaxial relationship derived from XRD measurements for (1122) GaN on m-plane sapphire.

Figure 36. Plan view SEM images of GaN samples grown on m-plane sapphire using an NH₃ flow rate of (a) 550 sccm, (b) 7060 sccm for the first 10 min and 550 sccm for the rest of the growth. The arrows indicate the c-axis of sapphire. (c) A 3D AFM image (3 x 3 μm²) of a V-shaped surface feature (Δz = 150 nm).

Figure 37. (a) Plan-view, (b) cross-sectional SEM, and (c) cross-sectional TEM images of GaN ELO sample with SiO₂ stripes oriented along the a-axis of sapphire. (d) Plan-view, (e) cross-sectional SEM, and (c) cross-sectional TEM images of the GaN ELO sample with SiO₂ stripes oriented along the c-axis of sapphire.

Figure 38. Extracted IQE values for the m- and c-plane LEDs on bulk GaN substrates using excitation intensity dependent PL measurements. The two LEDs have the same structure: 6 period 2 nm In₀.1₄Ga₀.₈₆N quantum wells with 12 nm In₀.₀₁Ga₀.₉₉N barriers. During the calculation of carrier densities for the samples, the B coefficient was assumed to be 1×10⁻¹¹ cm³s⁻¹.

Figure 39. (a) Integrated EL intensity and (b) relative external quantum efficiency of the m-plane LED on freestanding GaN and the reference LED on c-plane bulk GaN as a function of pulsed injection current density (1 μs, 0.1 % duty cycle). Both samples have the same device structure (MQW active region with 2 nm In₀.1₄Ga₀.₈₆N quantum wells, 12 nm In₀.₀₁Ga₀.₉₉N barriers, and p-Al₀.₁₅Ga₀.₈₅N electron blocking layers).

Figure 40. m-plane non-polar LEDs emitting at (a) 400nm (near ultraviolet), (b) 440nm (blue) and (c) 490nm (blue-green) at DC current ~100 Acm⁻². The LEDs were grown on nonpolar m-plane GaN substrates.
Figure 41. Normalized EL intensity as a function of the polarization analyzer angle for $m$-plane LEDs (6 period 2 nm InGaN QWs and 3 nm InGaN barriers) with different peak emission wavelengths. The polarizer angles of 0° and 90° correspond to E⊥c and E∥c, respectively. ....... 77

Figure 42. EQE and integrated EL intensity of a typical InGaN LED under pulsed current condition (1% duty cycle, 1kHz). The EQE reaches its peak value at relatively low current density of 50Acm$^{-2}$, and then monotonically decreases with the increase of current density. At the current of 1400Acm$^{-2}$, the EQE drops to 60% of its peak value. In some LEDs, the magnitude of drop is even larger. (from a commercial company, $\lambda$=460nm) ................................................ 78

Figure 43. (a) Relative EQE of m-plane LEDs grown on freestanding m-plane (1 T00) GaN substrates with varying Al composition (15%, 8%, 0%) in the EBL layers, measured under pulsed current, 1 $\mu$s pulse width and 0.1 % duty cycle. The inset shows the current-voltage dependence for the LED with 15% Al in EBL. (b) The EL efficiency loss as a function of the external applied voltage across the p-n junction of the LEDs, assuming negligible electron overflow at the current density ($\sim$80 Acm$^{-2}$) corresponding to the peak efficiency for the LED with 15% Al in EBL, which is very reasonable according to our calculations. ......................... 88

Figure 44. Relative EQE of c-plane DH LEDs with and without the EBL layers. The LEDs were measured under pulsed current, 1 $\mu$s pulse width and 1 kHz frequency. The LEDs were grown on c-plane GaN templates on sapphire, and their active regions are composed of a 6 nm thick undoped In$_{0.15}$Ga$_{0.85}$N layer (emission wavelength, $\lambda$~410nm). ................................................... 89

Figure 45. Calculated energy distribution of electrons in the active region at 3 different junction temperatures: 500K, 700K and 1000K. A flat band diagram schematic is used just to show the barrier height of the p-GaN and for a comparative picture of the three distributions, which actually correspond to three different band diagrams. The integrated tail of the Fermi-Dirac distribution makes up only 0%, ~1% and ~11% of the total current at the junction temperatures of 500K, 700K and 1000K, respectively. ................................................................. 91

Figure 46. Schematic of electron overflow caused by ballistic or quasi-ballistic electron transport across the InGaN active region. The electrons gain a kinetic energy after being injected into InGaN, which equals to $E+E_c+qV(x)$. These hot electrons will either traverse the active region ballistically and quasi-ballistically, escape recombination inside InGaN, and contribute
the electron overflow current, or be thermalized and captured inside the active region through interactions with LO-phonons.

**Figure 47.** (a) Calculated ratio of the overflow electron current to the total current as a function of the EBL barrier height ($\phi_{EBL}$) in non-polar m-plane LEDs, assuming flat-band conditions in the active region (i.e. 0V net potential drop across the InGaN active region after the applied external voltage compensates the built-in potential, which is ~0.5V), corresponding to 3.8, 4.0, and 4.7 V externally applied bias for the LEDs with 15%, 8%, and 0% Al in the EBL, respectively. (b) Calculated ratio of overflow electron current to the total current as a function of the applied voltage (forward direction) across the m-plane LEDs with three types of EBLs: 0% Al, 8% Al and 15% Al. The symbols in (a) and (b) represent the calculated points whereas the lines are guides to the eye.

**Figure 48.** Calculated (overflow electron current / total electron current) as a function of the applied forward voltage on the LED without staircase and without EBL, and the LED with two-step InGaN staircase and without EBL. Symbols represent the calculated values and solid lines are guides to the eye. The inset shows the schematics for the LED with and without (dashed line) the SEI. For simplicity, a one-layer SEI (one In$_{0.10}$Ga$_{0.90}$N intermediate layer) was employed for the calculations.

**Figure 49.** A schematic for the conduction band of a LED with a one-intermediate layer SEI (of thickness d and step height $\Delta E_c$). After being injected into the SEI from the n-GaN region, some electrons will have ballistic and quasi-ballistic (only one phonon emission or absorption) transport through the SEI [process a)], while the others (experiencing two or more scattering events) are considered to be thermalized in the SEI [process b)]. Under high bias condition and with a small step height $\Delta E_c$, the thermalized electrons from process b) might contribute to the overflow current through ballistic or quasi-ballistic transport [process c)]. For the calculations, the conduction band discontinuity between the active region and p-GaN is assumed to be 0.5eV, and no EBL is employed.

**Figure 50.** Schematic for a one-layer SEI and a two-layer SEI. For the one-layer SEI, the total SEI thickness is 9nm and the step height is 0.3eV. For the two-layer SEI, each step height is 0.15eV and each layer thickness is 4.5nm.
Figure 51. Calculated conduction band structures of c-plane (solid line) and m-plane (dotted line) InGaN LEDs with a 6nm In$_{0.15}$Ga$_{0.85}$N active region and a 10nm Al$_{0.15}$Ga$_{0.85}$N EBL. The injection current density for both LEDs is 1200Acm$^{-2}$. .............................................................. 111

Figure 52. Calculated electron overflow current as a function of the applied voltage on the LED for c-plane DH LEDs (6nm In$_{0.15}$Ga$_{0.85}$N active region) with different EBL heights (0% Al and 15% Al, respectively) and without SEI as well as a c-plane DH LED with SEI and without any EBL. ............................................................................................................................................ 112

Figure 53. (a) Schematic for the two m-plane LEDs with InGaN SEI before the active regions (to thermalize the injected electrons from the n-GaN layers), one of which has a 10nm EBL with 15% Al and the other one without any EBL in the p-region. (b) Relative EQE of the two m-plane LEDs with staircase electron injector: one with and one without EBL, respectively. The LEDs were measured under pulsed current with a frequency of 1kHz and 1µs duty width. ................ 115

Figure 54. Relative EQE of two c-plane LEDs with SEI inserted under the active region: one with and one without EBLs, respectively. The LEDs were measured under pulsed current with a frequency of 1kHz and 1µs pulse width. The SEI includes three intermediate InGaN layers with In composition of 3%, 6% and 10%, respectively. ..................................................................... 117

Figure 55. (a) A plan-view SEM image of a selectively-grown 40.5 pair Al$_{0.46}$Ga$_{0.54}$N/GaN DBR, with a pattern size of 150×150 µm$^2$. Each square is separated from each other by a 10 µm wide SiO$_2$ mask. DBR Surface is crack-free across the whole 2 inch wafer. (b) A close-up plan-view SEM image of the DBR sample near the SiO$_2$ mask at the pattern edge. (c) A cross-sectional SEM image of the selectively grown DBR sample showing individual GaN (dark) and AlGaN (bright) layers in the DBR. .......................................................................................................... 127

Figure 56. XRD ω-2θ scan of the AlGaN/GaN DBR with pattern size of 150 × 150 µm$^2$. From the clearly observed fringes the DBR pair thickness was determined as ~94 nm. ......................... 127

Figure 57. Reflectivity spectra from the DBR samples with different pattern sizes. With the decrease of pattern size, the stop bands blue shift, suggesting higher growth rate of DBR due to the selective growth. .................................................................................................................................. 129

Figure 58. Schematic for a full micro-cavity structure on patterned a 40.5 pair patterned Al$_{0.46}$Ga$_{0.54}$N/GaN DBR. ............................................................................................................. 129
**Figure 59.** Room-temperature micro-photoluminescence spectra (solid line) from full-cavity structure measured from the back surface of the sample. The dashed line represents the reflectivity data from the bottom DBR, which is a 40.5 pair patterned Al_{0.46}Ga_{0.54}N/GaN DBR. The top DBR is a 13 pair SiO_{2}/SiNₓ DBR, which has a peak reflectivity of ~99.5 %. The sample was excited by a frequency-doubled Ti:Sapphire laser emitting at 380 nm from the top DBR side. The device pattern size is 150 × 150 μm². The Q-value deduced from the PL spectrum (√λ/Δλ) is approximately 300.

**Figure 60.** A schematic for Mg δ-doping. The doping parameters include the nominally undoped GaN layer thickness d, and Mg flux P_{Mg} (determined by the Cp₂Mg flow rate f_{Mg} and deposition time t_{Mg}) with NH₃ flow at the same time. (after Ref. 137)

**Figure 61.** Curve fitting results of the generation rate G as a function of integrated PL intensity I_{PL} for a c-plane LED active layer (with 6 nm InGaN DH) on sapphire. The red curve represents the fits obtained using Equation 25. (Fitting parameters: P₁=8.59437×10^{23} and P₂=1.48422×10^{22}, goodness of fitting R²= 0.99863).

**Figure 62.** The internal quantum efficiencies of a c-plane LED active layer (with 6 nm InGaN DH) on sapphire, extracted from the excitation dependence of the PL intensity using a Ti:sapphire laser (370nm).
Abstract

General lighting with InGaN light emitting diodes (LEDs) as light sources is of particular interest in terms of energy savings and related environmental benefits due to high lighting efficiency, long lifetime, and Hg-free nature. Incandescent and fluorescent light sources are used for general lighting almost everywhere. But their lighting efficiency is very limited: only 20-30 lm/W for incandescent lighting bulb, approximately 100 lm/W for fluorescent lighting. State-of-the-art InGaN LEDs with a luminous efficacy of over 200 lm/W at room temperature have been reported. However, the goal of replacing the incandescent and fluorescent lights with InGaN LEDs is still elusive since their lighting efficiency decreases substantially when the injection current increases beyond certain values (typically 10-50 Acm⁻²). In order to improve the electroluminescence (EL) performance at high currents for InGaN LEDs, two approaches have been undertaken in this thesis. First, we explored the preparation and characterization of non-polar and semi-polar GaN substrates (including a-plane, m-plane and semi-polar planes). These substrates serve as promising alternatives to the commonly used c-plane, with the benefit of a reduced polarization-induced electric field and therefore higher quantum efficiency. It is demonstrated that LEDs on m-plane GaN substrates have inherently higher EL quantum efficiency and better efficiency retention ability at high injection currents than their c-plane counterparts.

Secondly, from a device structure level, we explored the possible origins of the EL efficiency degradation at high currents in InGaN LEDs and investigated the effect of hot electrons on EL of LEDs by varying the barrier height of electron blocking layer. A first-order theoretical model is proposed to explain the effect of electron overflow caused by hot electron
transport across the LED active region on LED EL performance. The calculation results are in agreement with experimental observations. Furthermore, a novel structure called a “staircase electron injector” (SEI) is demonstrated to effectively thermalize hot electrons, thereby reducing the reduction of EL efficiency due to electron overflow. The SEI features several $\text{In}_y\text{Ga}_{1-y}\text{N}$ layers, with their In fraction $(y)$ increasing in a stepwise manner, starting with a low value at the first step near the junction with n-GaN.
Chapter 1. Introduction

Lighting constitutes more than 20% of the total U.S. electricity consumption, a similar percentage in the European Union, and an even higher percentile in many developing countries.\(^1\) Power generation needed for lighting contributes approximately 1900 Mt CO\(_2\) per year (equivalent to 70% of the emissions from the world’s light passenger vehicles) according to the International Energy Agency (IEA)\(^2\). Compared to conventional lighting technology, light emitting diode (LED) lighting offers great promise as an energy-efficient, environment-friendly and affordable source of reasonably color-balanced white light owing to highly efficient LEDs. The performance of InGaN LEDs either as direct source of light generation and or pump source for relevant dyes is front and central to general lighting applications. Over the past two decades, the performance of InGaN-based LEDs has improved significantly and LEDs are now being put to use in applications such as general lighting (both indoors and outdoors), backlight for flat displays including flat panel energy efficient TVs. Among these applications, the general lighting application of the LEDs are particularly important market for InGaN LEDs, and it is expected in the near future the incandescent light sources and fluorescent light sources will be all replaced by the LED-based solid state lighting. The above mentioned applications promises an enormous market and huge economic values, and are the driving force for the ongoing research and rapid development in the area of LEDs.

The red, orange, and yellow LEDs are mainly made from AlGaInP materials system. For red LEDs, the active region is usually composed of Ga\(_{0.5}\)In\(_{0.5}\)P, which is lattice matched to GaAs substrates and has an energy bandgap of 1.9eV (650nm). The addition of Al to the GaInP materials can shift the emission wavelength towards shorter wavelengths (orange and yellow).
InGaN materials have been used for producing LEDs emitting ultraviolet, violet, blue and green light sources. The main attention of this work will be focused on the InGaN LEDs. So far enormous development has been achieved for the efficiency of the InGaAs LEDs in the recent decades. The state-of-the-art InGaN LEDs with a luminous efficacy of over 200 lm/W at room temperature have been reported\(^3\), as compared to typically 20-30 lm/W of incandescent light bulbs and 70-90 lm/W of fluorescent light sources.

However there are a few issues to address for the InGaN LEDs to be widely used in general lighting, especially where high luminescence with high injection current are required. One of the issues is that the InGaN-based LEDs suffer from loss of efficiency at high injection levels needed, which has been dubbed as the “efficiency droop”.\(^4\) This manifests itself as the external quantum efficiency (EQE) reaching its peak value at current densities even lower than 50 A/cm\(^2\) and monotonically decreasing from there on with further increase in current.

The major goal of this work is to understand the underlying mechanism of the so-called “efficiency droop” issue, and to achieve an optimum LED structure design to improve the EL efficiency of the InGaN LEDs at high injection currents. In the following sections, the background for this thesis will be presented.

### 1.1 InGaN light emitting diode (LED) structure

All LED structures were grown on either GaN/sapphire template or bulk GaN in a vertical low-pressure metalorganic chemical vapor deposition (MOCVD) system. As shown in Figure 1, the epitaxy of an InGaN LED starts with an n-type GaN which is doped with Si using SiH\(_4\) as dopant source. It is followed by an underlayer (60 nm Si-doped In\(_{0.01}\)Ga\(_{0.99}\)N, for example) just beneath the active region (6-period multiple quantum wells as an example in the
figure) for improved quality. An AlGaN electron blocking layer is deposited on top of the active quantum well region to reduce the outgoing electrons from the active region. A ~100 nm Mg-doped p-GaN is deposited on top of the AlGaN layer to serve as the source for hole injection. A simplified conduction band profile is shown on the right side of Figure 1. After mesa (250 μm diameter) etching, Ti/Al/Ni/Au (30/100/40/50 nm) metallization annealed at 800 °C for 60 seconds was used for n-type ohmic contacts and 5 nm/5 nm Ni/Au contacts were used for the semi-transparent p-contacts. Finally, 30/50 nm Ni/Au contact pads were deposited on the top of part of the mesa.

Figure 1. Schematic for a typical InGaN LED structure in this thesis with a simplified conduction band edge profile shown on the right side.

1.2 Carrier recombination in LEDs

As mentioned earlier, InGaN LEDs are composed of n-type region, active region and p-type region, etc. The electrons and holes are injected from n-side and p-side by drift-diffusion current into the active region. The recombination of electrons and holes within the active region
could be radiative or non-radiative. The radiative recombination rate is described as $B\Delta n^2$, where $B$ is the radiative recombination coefficient, $\Delta n$ is the excess electron density, assuming $\Delta n$ is much larger than the intrinsic carrier density since GaN is a large band-gap material and has a low intrinsic carrier density. At low injection currents, the major nonradiative recombination process is Shockley-Read-Hall (SRH), with its recombination rate described as $A\Delta n$, where $A$ is the SRH recombination coefficient, $n$ is the electron density. Another type of nonradiative recombination is Auger recombination, in which the energy given off by the recombination of electron and hole is used to excite another carrier to a higher energy which in turn thermalizes down to lower energy state by phonon emission. The Auger recombination rate is given by $C\Delta n^3$. The Auger recombination will be covered in details in section 4.3, Chapter 4. Another form of carrier loss mechanism is carrier overflow or spillover, especially for electrons as they are lighter in terms of effective mass. The electrons could escape the active region, ending up with recombining at the p-region or the contact and not contributing to the desired emission in the active region. In order to have high internal quantum efficiency for the LEDs, both nonradiative recombination and carrier overflow or spillover should be minimized.

1.3 The motivation of non-polar nitrides for light emitting applications

In order to achieve high IQE for LEDs, one promising way is to use non-polar GaN as the substrate for LEDs. Currently almost all research about GaN is about c-axis oriented wurtzite epilayers. In this kind of GaN-based heterostructures, the internal spontaneous and piezoelectronic polarization effects can cause strong electric field in the nitrides interface. Although this electric field can be advantageous for formation of two-dimensional electron gas
in field-effect transistors (FETs), it can also cause spatial separation of electrons and holes in quantum wells of InGaN LEDs, thereby increasing the radiative lifetime and hence reducing the quantum efficiency, and can also cause the red shift of LED emission (see Figure 2). This is called Quantum Confined Stark effect.

![Figure 2. Calculated band profiles in (5nm GaN)/(10nm Al_{0.1}Ga_{0.9}N) quantum wells by self-consistent effective mass Schrödinger-Poisson calculations. a, The very large electrostatic fields in the [0001] orientation result in a quantum confined Stark effect and poor electron-hole overlap. b, The [1100] orientation is free of electrostatic fields, thus true flat-band conditions are established.](image)

There are two ways to solve this polarization issue. One is to grow cubic rather than hexagonal GaN, which is nonpolar along cubic [001] direction, and therefore can avoid strong polarization-induced electric field in heterointerfaces. Unfortunately, cubic GaN is metastable and it is also very difficult to be achieved. The other way is to grow a- or m-plane hexagonal GaN, rather than c-plane GaN. These kinds of GaN can be called nonpolar GaN, since the c axis is parallel to the substrate surface. For this kind of oriented GaN, there is no polarization-induced...
electric field in nitrides interfaces. Figure 3 shows schematically the m-, a-, and c-plane of GaN. These planes are perpendicular to each other.

Figure 3. Schematic representation of m-, a-, r- and c-plane of GaN.

1.4 Materials growth procedures – metalorganic chemical vapor deposition (MOCVD)

MOCVD is an epitaxial growth method for preparation of semiconductor thin films-based devices. The MOCVD technology has been established its ability for mass production of high-quality epitaxial layers, and has been widely used for producing nitride-based LEDs, and other compound semiconductor-based devices (such as AlInGaP-based LEDs). The growth of epitaxial layers is typically achieved by introducing alkyls for group III, and hydrides for group V onto a heated substrate inside a vacuum chamber. For GaN system, trimethylgallium (TMGa) or triethylgallium (TEGa) is used as Ga source, and NH₃ gas is used as N source. Additionally,
trimethylindium (TMIn), and trimethylaluminium (TMAI), diluted SiH₄ gas are used as In, Al, and Si (n-type dopant) sources.

The flow rate of each metalorganic (MO) source is controlled by three factors (as shown in Figure 4): MO source temperature (source temperature determines MO vapor pressure, \( P_{MO} \)) \((T)\), carrier gas flow rate \((f_{carrier})\), and bubbler pressure \((P_{bubbler})\):

\[
f_{MO}(mol/\text{min}) = \frac{f_{carrier}(sccm/\text{min})}{22414(sccm/mol)} \cdot \frac{P_{MO}(T)}{P_{bubbler} - P_{MO}(T)}
\]

Equation 1

where the MO source vapor pressure \( P_{MO} \) is determined by \( T \) through:

\[
\log[P_{MO}] = B - \frac{A}{T}
\]

Equation 2

where \( A \) and \( B \) are constants determined empirically for each specific source.

![Figure 4](image)

Figure 4. A schematic for MO source bubbler used in MOCVD system for providing MO source. The MO source mole flow rate is determined by (1) MO source temperature, (2) carrier gas flow rate, and (3) bubbler pressure (controlled by an upstream pressure controller).

1.5 Epitaxial lateral overgrowth (ELO)

In order to improve the materials properties, epitaxial lateral overgrowth (ELO) method has been used often in MOCVD system to reduce the defect density. The basic idea of this
growth method is to filter out the defects by employing the dielectric masks (such as SiO$_2$, or SiN$_x$), as shown schematically in Figure 5. First, planar GaN template is grown on substrates such as sapphire, SiC or Si, which is followed by deposition of a dielectric (SiO$_2$ or SiN$_x$) mask by PECVD. By using standard photolithography, a set of parallel stripes is defined on the GaN template, with the separations between the dielectric stripes called “window”, as shown in Figure 5(a). Then the patterned sample is loaded back into the MOCVD system for GaN regrowth. During this procedure, epitaxial growth of GaN will start in the window regions, where the microstructure of the underlying GaN template is reproduced, whereas no growth will occur above the masked area [Figure 5(b)]. The overgrown GaN will extend both vertically or laterally whose ratio highly depends on the growth conditions used. Laterally grown regions are called “wing” region, which contains much fewer defects. After certain amount of regrowth time, the overgrown GaN will get coalesced, and a few defects will be generated at the coalescence boundary [Figure 5(c)]. A thorough review about this growth technique could be found in refs. 9,10.

In c-plane GaN ELO case, the stripes of dielectric films are usually aligned along the GaN m-axis [1̅00] so that the laterally overgrown wings expand along the GaN a-axis [11̅20], as shown in Figure 5(d). It has been demonstrated that when the mask stripes are aligned along this direction, the ratio of lateral growth rate to vertical growth rate will be higher and the sidewall facet is easier to be controlled by growth conditions.$^{11}$ In contrast, when the mask stripes are aligned along the GaN a-axis [11̅20] and the lateral growth will advance along the GaN m-axis, the lateral overgrowth will be limited by a slow growth rate of (1̅01) facets, which are the most stable facets in GaN and difficult to be eliminated.$^{11}$ However, in the (11̅20)a-plane
or (1T00) m-plane GaN ELO cases, the stripe-shaped masks should be aligned along the GaN m-axis [1T00] and the GaN a-axis [1120], so that the overgrown wings advance laterally along the GaN c-axis and result in an effective defect reduction within the overgrown GaN wing regions, as will be shown in chapter 2.

Figure 5. Schematic for an ELO process: (a) stripe-shaped mask (typically SiO₂ or SiNₓ dielectric films) on planar GaN template was obtained through photolithography procedures with the open area ("window") exposed for the regrowth process. (b) During regrowth step, the GaN material starts growth at the window regions, and expands both vertically and laterally. In contrast to the window regions, the overgrowth GaN above the mask regions which is normally called "wings" has very few defects. (c) After certain amount of time, the GaN wings will coalesce and form continuous films. A few defects will be generated at the coalescence boundary, where the two opposite wings coalesce. (d) The directions for the patterning of ELO mask for c-plane, a-plane and m-plane GaN.
ELO. It should be noted that in a-plane and m-plane GaN ELO, the Ga-polar wings have higher lateral growth rate than that of N-polar wings. So the coalescence boundary is not in the middle of the masked regions.

The ELO technique has resulted in a significant improvement in laser diode (LD) lifetimes (>10,000 hours) at room temperature,\textsuperscript{12} but it requires extra procedure for preparing dielectric mask \textit{ex-situ}, which costs time and possible sample contaminations during loading and unloading. Another important ELO technique “nano-ELO” has been reported by Xie et al,\textsuperscript{13} which utilizes \textit{in-situ} grown porous SiN\textsubscript{x} film in MOCVD as the growth mask. However since the growth windows (i.e., nano-sized pores in SiN\textsubscript{x} film) is formed randomly during the \textit{in-situ} SiN\textsubscript{x} deposition, this technique could not define the GaN lateral growth directions, which is important for an effective defect reduction in non-polar GaN materials.

1.6 Methods for measurement of LED internal quantum efficiency (IQE)

The performance of LEDs could be evaluated by internal quantum efficiency (IQE), external quantum efficiency (EQE), power conversion efficiency (or called “wallplug efficiency”) and luminous efficacy. The EQE is related to the IQE by extraction efficiency (EXE): \( \text{EQE}=\text{IQE} \times \text{EXE} \). The power conversion efficiency is defined as \( \text{P}_{\text{opt}}/(V_f \times I_f) \), where \( \text{P}_{\text{opt}} \) is the output optical power of the LED, \( V_f \) forward voltage, \( I_f \) the forward current. Luminous efficacy (\( \eta_L \)) is often defined as \( \Phi_L/(V_f \times I_f) \), where \( \Phi_L \) is the photometric photon flux (luminous flux). Power conversion efficiency (PCE) is related to the luminous efficacy (\( \eta_L \)) by \( \eta_L=\text{PCE} \times V_\lambda \), where \( V_\lambda \) is the wavelength-dependent photo-responsivity of the human eye expressed in terms of lm/W. The EQE, PCE, and \( \eta_L \) could be measured directly. The extraction efficiency could be calculated by using ray-trace modeling and also could be estimated experimentally by using temperature
dependent EL method. With the decrease of temperature, the EQE from EL measurement will reach its maximum due to the increase of IQE. At low temperature (e.g. 7K in our case), with the IQE assumed to the 100% (which is not necessarily true, no direct evidence), the up limit of EXE could be approximately determined. As one of the most important figures of merit of a LED chip, IQE, which is defined as the ratio of the total number of the photons generated inside the LED structure to the total number of carriers injected either electrically or optically, could be obtained by:  

- temperature-dependent PL measurement, or
- excitation power dependent-PL measurement.

For the first method, as the sample temperature is decreased, the nonradiative recombination lifetime increases and the peak IQE can be considered close to 100% at low temperature (e.g. 7K). At each temperature step, the PL intensity is measured as a function of the excitation power. By normalizing the peak relative EQE (the ratio of PL intensity to the laser power) at room temperature to the peak relative EQE measured at 7K, IQE values could be obtained.\(^\text{14}\) It should be noted that the IQE highly depends on the excitation power therefore the generated carrier density especially at low excitation power region. So the excitation laser wavelength should be tuned to avoid the excitation of the top GaN or the barriers inside the active region. It should be noted that the accuracy of the IQE values obtained with this method is dependent on the assumption that the peak IQE value at low temperature is unity, which is not necessarily true, and has no direct evidence.

The second method is the excitation power-dependent PL. In our case, a resonant excitation laser source is used for the LED samples at room temperature using a frequency-doubled 80 MHz repetition rate femtosecond Ti:Sapphire laser with 100 fs pulses. The excitation laser wavelength was 370 nm, below the bandgap of the quantum barriers and top GaN. As such,
the photo-excited electron-hole pairs can only be generated within the quantum wells, thereby avoiding optical carrier generation in the barriers and also carrier injection effect which has been observed to cause efficiency droop in the electroluminescence (EL) case. During PL measurements, the average excitation power density was varied from 7 to 280 W/cm² using neutral density filters.

In order to measure the IQE with the power-dependent PL, a similar procedure to the one described in Ref. has been used. In our model we assume that at steady state the total generation rate \( G \) is equal to the total recombination rate \( R \) which includes Shockley-Read-Hall nonradiative recombination \( (A_n) \), bimolecular radiative recombination \( (Bn^2) \), and Auger recombination \( (Cn^3) \) if any, where \( n \) is the carrier density. The measured PL intensity could be represented as \( I_{PL} = \eta_c B n^2 \), where \( I_{PL} \) is the integrated PL intensity, the collection factor \( \eta_c \) includes escape efficiency of photons as well as the collection efficiency of luminescence by the optics/detector, which is constant during a given measurement but different from measurement to measurement even though attempts are made to keep the collection geometry the same. This variability does not, however, affect the values of the efficiencies reported here.

From the above discussion it follows that the total generation rate at steady state is

\[
G = \frac{A}{\sqrt{\eta_c B}} \sqrt{I_{PL}} + \frac{1}{\eta_c} I_{PL} + \frac{C}{(\eta_c B)^{3/2}} \left( \sqrt{I_{PL}} \right)^{3/2}
\]

Equation 3

which can also be calculated separately from excitation laser power as described in Ref. 16 by

\[
G = P_{laser} (1 - R) \alpha / (A_{spot} \nu)
\]

Equation 4

where \( P_{laser} \) is the laser power, \( R \) is the Fresnel reflectivity at the sample surface, \( \alpha \) is the absorption coefficient of the quantum wells at the laser emission wavelength, \( \nu \) is the photon
energy from laser. By fitting the calculated generation rate as a function of the square root of integrated PL intensity \( \sqrt{I_{PL}} \), the three coefficients for each term in Equation 3 can be obtained, from which the IQE = \( B n^2 / G = (I_{PL} / \eta_c) / G \) values are subsequently calculated. Knowing the \( B \) value, one can also obtain \( A \) and \( C \) recombination parameters [see Appendix A for detailed procedures]. However, it should be mentioned here that the IQE does not depend on the assumed value of the \( B \) parameter (\( 1 \times 10^{-11} \) cm\(^3\)s\(^{-1}\) used here). The downside of this method is that it has assumed a constant \( B \) parameter with the increase of laser excitation power. However, this assumption is not true due to two reasons: i) in \( c \)-plane LEDs the screening of polarization-induced electric field with the increase of injected carriers could change the radiative recombination coefficient \( B \); ii) the formation and dissociation of excitons cannot be neglected even at room temperature given an exciton binding energy of the order 20 meV,\(^1\)\(^7\),\(^1\)\(^8\) also leading to a strong change in \( B \) with injected carrier density inside the active region.\(^1\)\(^4\) Yet, this excitation-power dependent PL method could still be used to make a wafer-to-wafer comparison to a first extent. In the following discussion, the IQE values were obtained by using one the above two methods.

### 1.7 Organization of this thesis

As we discussed above, the major goal of this work is to improve the LED performance at high current. So our study begins with the development of non-polar GaN materials, which is beneficial for obtaining high-efficiency InGaN LEDs due to the elimination of polarization-induced electric field. In Chapter 2, we will describe the epitaxial growth of high-quality non-polar \( a \)-plane and \( m \)-plane GaN materials with epitaxial lateral overgrowth method. Chapter 3 will focus on the comparison of the performance of InGaN LEDs on \( c \)-plane and non-polar
planes (m- and a- planes). Chapter 4 will focus on the efficiency droop issue in InGaN LEDs in both polar and non-polar orientations. We will discuss the possible mechanisms responsible for the efficiency loss at high current, including Auger non-radiative recombination, electron overflow, etc. Among all of these mechanisms, we propose that electron overflow is the dominant reason for the efficiency loss issue at high current levels. Both theoretical models and experimental data will be used to explain the effect of the electron overflow on the LED EL efficiency. A novel structure which is used to reduce the electron overflow caused EL efficiency loss is proposed. In Chapter 5, we will present the results of the preparation of highly reflective and crack-free nitride distributed Bragg reflectors (DBRs) and their applications to vertical cavity surface emitting lasers (VCSELs). Finally conclusions and suggestions for future work will be presented in Chapter 6.
Chapter 2. Epitaxial growth of non-polar GaN materials

Non-polar GaN materials are beneficial for obtaining high internal quantum efficiency due to their polarization-free nature. However, the growth conditions for the non-polar GaN materials are different from those of c-plane GaN due to the different crystalline orientations. In this chapter, we will first present the growth conditions for a-plane GaN on r-plane sapphire. Next, epitaxial lateral overgrowth technique is employed to improve the materials quality of a-plane GaN, which is evidenced by structural and optical measurement results. In addition to a-plane orientation, we also explored the other orientation of non-polar GaN: m-plane GaN. m-plane GaN is well-suited for optoelectronic applications in that it is predicted to have a reduced valence band effective mass, and thus smaller acceptor binding energy, and larger optical matrix elements relative to its c-plane counterpart, thereby improving the performance of LEDs and LDs.¹⁹

2.1 Epitaxial growth of a-plane GaN on r-plane sapphire

(1120) a-plane GaN films were grown on (1 T02) r-plane sapphire (with ± 2° surface tolerance). Prior to growth, the sapphire substrates were annealed at 1400°C for one hour in air ambient with annealing furnace to generate atomic steps on substrate surface. After chemical cleaning, sapphire substrates, one at a time, were loaded into the low-pressure custom-designed Organo-Metallic Vapor Phase Epitaxy (OMVPE) system. Following in-situ annealing of sapphire, a low-temperature (LT) GaN buffer layer was deposited at 550°C with a thickness of 45nm. In the case of using high-temperature (HT) AlN as buffer layer, the buffer layer growth temperature and thickness were 1080°C and around 100nm, respectively. After buffer layer
growth, the temperature was raised to a value in between 1040°C and 1070°C, and a-plane GaN epilayer was grown subsequently. Trimethylgallium (TMGa), trimethylaluminum (TMAI) and ammonia were used as the Ga, Al and N sources, respectively. The as-grown samples were characterized by Scanning Electron Microscopy (SEM), Atomic Force Microscopy (AFM), and high-resolution x-ray diffraction (XRD).

The growth parameter space for optimization included chamber pressure, ammonia flow rate, and epilayer growth temperature. Figure 6 shows the effect of growth pressure on a-GaN surface morphology. During optimization, 0.9 μm-thick GaN films were grown under two different growth pressures: 30 torr and 200 torr. As shown in Figure 6(b), the lateral growth rate under 200 torr was rather small with respect to the vertical growth rate, and consequently we could hardly observe the presence of flat fully coalesced area on the sample surface. By comparison, a-plane GaN grown under 30 torr [Figure 6(a)] showed a better tendency of lateral growth, albeit with large pits decorating the surface. The pits were very stable under unoptimized growth conditions, and could not be eliminated by simply increasing the film thickness. Wu et al. reported that these triangular pits were composed of one \{0001\} and two \{10 \bar{1} 1\} facets.

Figure 6. SEM images of 0.9μm (1\bar{1}2\bar{0}) a-plane GaN grown on (1 \bar{1}02) r-plane sapphire at 980°C, with 7000ccm NH₃, under (a) 30torr, (b) 200torr, respectively.
Ammonia flow rate is also an important parameter for growth of a-plane GaN. Figure 7 shows the effect of NH₃ flow rate on the surface morphology of a-plane GaN. The three samples shown here have the same thickness of around 1.5μm, with different sized pits on the surface. With decreasing flow rate of NH₃, the size of the triangular pits was reduced from several microns to zero. Therefore, a relatively low V/III ratio was found to be beneficial in reducing the size of pits on the sample surface and attaining coalesced GaN films. Additionally, we can determine [0001] or [000\overline{1}] direction according to stripes and the shape of triangular pit on surface²⁰, as shown in this figure.

![SEM images of (1\overline{1}20) a-plane GaN grown under 30torr, at 1070°C, with NH₃ flow rates of (a) 7060ccm, (b) 4500ccm, (c) 1000ccm, respectively.](image)

We have also investigated the influence of the growth temperature on a-plane GaN, as shown in Figure 8. The pit size was reduced with increasing growth temperature from 1050°C to 1090°C. However, from these SEM images, we can also see that the surface undulation became
more significant with increasing growth temperature, which was also confirmed by surface roughness measurements using AFM which are not shown here. The FWHM value of the X-ray rocking curve of GaN (1120) showed in-plane anisotropy, and was a function of the in-plane azimuth angle, consistent with the report by H. M. Wang et al.\textsuperscript{21} The FWHM values of X-ray rocking curve were 0.46°-0.67°, 0.29°-0.43°, and 0.28°-0.43° for samples grown at 1050°C, 1070°C, and 1090°C, respectively. Higher growth temperature could yield a-plane GaN with improved crystalline quality. Therefore, the choice of optimum temperature should be a compromise of surface morphology and crystalline quality.

Figure 8. SEM images of (1120) a-plane GaN grown under 30torr with NH\textsubscript{3} flow rate of 1000 sccm, with growth temperature being (a) 1050°C, (b) 1070°C, (c) 1090°C, respectively.

Figure 9 shows the surface morphological evolution during early stage of a-plane GaN growth. These SEM images show that the 45nm LT-GaN buffer was featureless, as shown in Figure 9(a). Continuing on with further growth, the 20nm-thick GaN in Figure 9(b) was
dominated by nucleation islands. When the thickness increased to around 50nm (Figure 9(c)), the islands began to coalesce and form elongated features running along one direction. We determined this to be the c-direction of GaN. With the growth continuing, the elongated islands started to merge with each other, as shown in Figure 9(d). For the 300nm-thick GaN [Figure 9(e)], the sample was almost fully coalesced, with some small pits on the surface. When the thickness reached 1.5μm [Figure 9(f)], the film fully coalesced. From this image, a-plane GaN showed stripe features along the c-direction, and surface undulation along the m-direction ([1̅100]) as well, which is perpendicular to the c-direction [0001]. From the elongated island growth, we can also conclude that the growth rate along the c-direction is much higher than that along the m-direction. It is worth mentioning here that by performing lateral overgrowth using our optimized growth conditions, the ratio of the lateral growth rate along the c-direction to the vertical growth rate was around 3-4. Additionally, from the growth evolution, it is suggested that at the very beginning of a-plane GaN epitaxy, the growth follows a Volmer-Weber (VW) mode, which gives way to a Frank-van der Merwe (FM) mode after most of the islands have merged to each other.
Figure 9. Surface morphological evolution during early stage of a-plane GaN growth: (a) LT-GaN buffer, (b) 20nm GaN, (c) 50nm GaN, (d) 100nm GaN, (e) 300nm GaN, (f) 1.5μm fully coalesced GaN. Note that c direction is the stripe direction shown in (f), and all samples here have their c directions parallel to each other.

In order to further optimize a-plane GaN films, we have also investigated the effect of HT-AlN buffer layers in addition to LT-GaN buffers, which was used for the above-mentioned a-plane GaN samples. The AlN buffer layer was 100nm thick, and was grown at 1080°C using TMAI as the Al source. The flow rates for TMAI, and NH₃ were 10μmol/min, and 7ccm, respectively. A-plane GaN epilayers were grown at 1070°C, 30torr, with 1000ccm NH₃. XRD measurements with 2θ-ω scan (see Figure 10) showed (2204) r-plane of sapphire, (1120) of AlN and (1120) of GaN parallel to each other. The full width at half maximum (FWHM) value of the x-ray rocking curve for (1120) GaN was in the range of 0.29°-0.52°. AFM and SEM measurements were used to characterize the surface morphology of these a-plane GaN films, the results of which are shown in Figure 11. AFM root mean square (RMS) roughness was about 15nm, similar to that of a-plane GaN grown at the same temperature using a LT-GaN buffer. An
SEM image revealed fully coalesced surface and stripe features along the c-direction, which was also observed in the case of LT-GaN buffer layers.

Figure 10. XRD spectra using 2θ-ω scan for a-plane GaN grown with LT-AlN buffer layer on (1102) r-plane sapphire.

Figure 11. Surface morphology measurements of (1120) a-plane GaN films grown with HT-AlN buffer, by (a) AFM and (b) SEM measurements.

From the above experimental results, stripes features along the c-direction and surface undulation along the m-direction were found on all a-plane GaN samples, no matter what kind of buffer layer was used for epitaxy. First, we can rule out the possibility that the surface undulation
comes from the atomic steps of annealed r-plane sapphire formed during annealing as the same surface features were obtained on unannealed sapphire substrates, which does not have those atomic steps on its surface. One possible explanation is that the surface undulation may be caused by the disparity of migration lengths of adatoms along the two perpendicular directions, c- and m-directions, during growth, as shown in the case of high-index GaAs surfaces such as GaAs (331)\(^2\). This disparity of migration length could be enhanced with increasing temperature, therefore resulting in more significant surface undulation for a-plane GaN films at elevated growth temperature.

In summary, a-plane GaN growth conditions by MOCVD, including the chamber pressure, ammonia flow rate and epilayer growth temperature, have been optimized using LT-GaN buffer layer. Low pressure, low ammonia flow rate (namely low V/III ratio), high growth temperatures have been found to be beneficial for the growth of fully coalesced and relatively higher crystalline-quality a-plane GaN films. We also explored a-plane GaN epilayer using HT-AlN buffer layer, with similar crystalline quality and surface morphology to a-plane GaN using LT-GaN buffer.

### 2.2 Epitaxial lateral overgrowth of a-plane GaN on r-plane sapphire

Based on the growth parameter optimization in the previous section about planar a-GaN, we will go a step further, and use the epitaxial lateral overgrowth (ELO) method\(^2\) to reduce the density of threading dislocations in a-plane GaN using MOCVD.\(^2\),\(^2\) However, one must consider the wing tilt, which has been shown to be an important factor also for c-plane GaN ELO.\(^2\) In this section, we investigate the effects of MOCVD growth parameters on the overgrown a-plane GaN material quality inclusive of the wing tilt, structural, and optical properties.
2.2a Experimental procedures

The (1120) \( a \)-plane GaN films investigated in this study were grown on (1\( \bar{1} \)02) \( r \)-plane sapphire substrates. For the ELO, 1.5 \( \mu \)m-thick \( a \)-plane GaN templates with low-temperature grown GaN nucleation layers were used. A SiO\(_2\) layer approximately 100 nm thick was grown on the \( a \)-plane GaN template by remote plasma enhanced chemical vapor deposition (PECVD). A striped mask pattern was transferred onto SiO\(_2\)/GaN/sapphire using conventional photolithography and buffered oxide etch (BOE). The pattern consisted of 4 \( \mu \)m-wide open windows and 10 \( \mu \)m- or 20 \( \mu \)m-wide SiO\(_2\) stripes that were oriented along the (1\( \bar{1} \)00) direction (i.e. the m-axis) of GaN that would cause the lateral growth fronts to advance along the \( c^+ \) and \( c^- \) directions. The patterned template was then reloaded into the MOCVD chamber for overgrowth. Three \( a \)-GaN ELO samples were investigated for this particular study, samples A, B, and C. Samples A and B were grown with trimethylgallium and NH\(_3\) flow rates of 157 \( \mu \)mol/min and 3000 sccm, respectively. Sample A was grown in a single step at 1050 \( ^\circ \)C for 3 h while sample B was grown in two stages: at 1000 \( ^\circ \)C for 2 h in stage I and at 1050 \( ^\circ \)C for 3 h in stage II. Sample C was grown using 560 sccm NH\(_3\) in the second stage with the rest of the conditions kept identical to those employed for sample B. All three samples were grown on two separate templates containing 10 \( \mu \)m- and 20 \( \mu \)m-wide SiO\(_2\) stripes placed side by side in the growth chamber.

The as-grown samples were characterized by SEM, high-resolution XRD, transmission electron microscopy (TEM), AFM, photoluminescence (PL), time-resolved PL (TRPL), and near field scanning optical microscopy (NSOM). Low-temperature PL was performed on GaN layers using 325 nm excitation from a HeCd laser. TRPL measurements were performed at room temperature using 325 nm excitation from a Ti:Sapphire oscillator/regenerative amplifier.
pumped optical parametric amplifier and a Hamamatsu streak camera. The excitation density was kept well below the stimulated emission threshold and the spot size (~350 μm diameter) was much larger compared to the wing/window widths. NSOM measurements were performed at room temperature in the illumination mode where a 325 nm HeCd laser was used for excitation through a metal-coated cantilevered optical fiber probe with a 350 nm aperture. NSOM PL intensity mapping was carried out using a photomultiplier tube to collect the overall PL spectrum with the scattered and reflected laser light blocked with an optical filter. The cross-sectional SEM measurements were performed on the 20 μm stripe samples for a better view of the overgrown layer dimensions. The rest of the analysis was focused on the 10 μm stripe samples unless otherwise indicated.

2.2b Scanning electron microscopy (SEM) images

The plan-view SEM image of sample A in Figure 12(a) shows that the a-plane GaN stripes have vertical (0001) and (000 \( \overline{1} \) ) side walls after 0.5 h of overgrowth with no other facets present. As shown in Figure 12(c) the surface of sample A was fully coalesced after a total of 3 h of overgrowth but with striations along and steps perpendicular to the c-axis. The cross-sectional SEM image [Figure 12(b)] clearly shows that the wings with Ga-polarity are 4-6 times wider than those with N-polarity. As a consequence of the inherent wing tilt and significantly different growth rates of the two opposing wings, a clear height difference appears at the coalescence front, as illustrated by the schematics in Figure 13(a) and (b). This wing-tilt-induced height difference causes a significant surface undulation in addition to defects at the coalescence fronts of a-plane GaN, and is the origin of the steps observed in Figure 12(c). However, the height difference is mainly a consequence of the difference in growth rates of the Ga- and N-polar wings. In order to
obtain uniform coalescence and smooth surface, this height difference should be decreased or even eliminated if possible by reducing the difference between the widths or growth rates of the two opposing wings.

Our experimental results suggest that the growth temperature is a highly effective parameter to control the difference in the growth rates of the Ga- and N-polar wings. Therefore, for sample B, a two-stage growth method was employed. A growth temperature of 1000 °C was used in stage I to favor vertical growth while maintaining a relatively low lateral growth rate which is not drastically different for the lateral Ga- and N-polar fronts at this temperature. Figure 12(d) shows that, after the stage I growth, the sidewalls of $a$-GaN stripes are composed of facets other than (0001) or (0001). It is noted that most of the lateral growth has been achieved with no coalescence during stage I where the Ga- to N- polar wing width ratio is close to 1.5-2. The growth temperature was elevated to 1050 °C in stage II to enhance the lateral growth for complete coalescence. A quite smooth surface was achieved after full coalescence, and the Ga- polar wing was only 1.5-2 times wider than the N-polar wing as shown in Figure 12(e) and (f). These results indicate that the uneven growth rates of Ga- and N-polar wings could be suppressed to reduce the step height at the meeting fronts by enhancing the vertical growth rate during the early stages of the growth.
Figure 12. Plan-view SEM images for sample A after (a) 0.5 h and (c) 3.0 h of growth. (b) Cross-sectional SEM image for sample A after 3 h of growth. (d) Cross-sectional SEM image for sample B after 2 h of growth at 1000 °C. (e) Cross-sectional and (f) plan-view SEM images for sample B after a total of 5 h of growth.
Figure 13. Schematics for a-plane GaN ELO showing the origin of the height difference between two neighboring wings (not in exact proportion), with Ga- to N- polar wing width ratios of (a) 5:1 and (b) 1.6:1. The larger this ratio, the larger the height difference is between the two opposite wings at the coalescence fronts. (c) Schematic showing the inclination of the growth planes due to the 1.05° miscut of the r-plane sapphire towards its [0001] c-axis. The miscut of the substrate, and therefore, the resultant inclination of the GaN growth planes is the origin of the elevated meeting fronts observed from AFM.

2.2c X-ray Diffraction (XRD) results

The wing tilt is an important issue inherent in conventional ELO. To determine the wing tilt angles of samples A and B, XRD rocking curve measurements were carried out with three different $\phi$ angles, where $\phi$ is the angle of rotation about the sample surface normal and is defined as 0° when the projection of incident x-ray beam is parallel to the SiO$_2$ mask stripes. As shown in Figure 14(a) and (b), both samples exhibited only one a-plane GaN diffraction peak when $\phi=0^\circ$, with a full width at half maximum (FWHM) of 0.40° and 0.19° for samples A and B, respectively. However, two and three peaks were observed for samples A and B, respectively, for
\( \phi = 90^\circ \) (the projection of the incident x-ray beam is perpendicular to the mask stripes), with the peak order reversed for \( \phi = 270^\circ \). The FWHM values for the strongest peaks are 0.44° and 0.40° for samples A and B, respectively. The strongest peak observed for sample A is from the Ga-polar wings as determined from the XRD geometry used, since the surface area of the Ga-polar wing is much larger than that from the combination of the N-polar wing and the window as verified from the SEM images discussed above. Therefore, the weak peak is assumed to be from the windows and/or N-polar wings. The separation between the two peaks indicates a tilt angle of 0.86°, which is much larger than that obtained from the large angle convergent beam electron diffraction (LACBED) (0.25°) (see Figure 15). This discrepancy may be attributed to the local nature of the LACBED measurement, while XRD characterizes a much larger area and provides a value averaged over a large area. As shown in Figure 14(b), sample B exhibits smaller XRD linewidths compared to sample A, which indicates improved crystalline quality. Moreover, peaks from all three ELO regions are distinguishable for this sample. The strongest peak in Figure 14(b) is from the Ga-polar wings (0.44° tilt), while the central one is from the crystal plane in the windows, and the third is from the N-polar wings (0.37° tilt).
Figure 14. XRD rocking curve data for (a) sample A and (b) sample B with different \( \phi \) angles. The dashed lines correspond to the multiple Gaussian fits to the rocking curve data.

2.2d Transmission electron microscopy (TEM) results

TEM studies indicated a significant reduction in threading dislocation densities by ELO. For sample A, the threading dislocation density was reduced from \( 4.2 \times 10^{10} \) cm\(^{-2} \) in the windows to \( 1.0 \times 10^{8} \) cm\(^{-2} \) in the wings, as evidenced in Figure 16(a). However, a relatively high density of
basal stacking faults (BSFs), $1.2 \times 10^4 \text{ cm}^{-1}$, is still present in the wing areas compared to $1.3 \times 10^6 \text{ cm}^{-1}$ in the windows, due to the low formation energy of stacking faults in the basal plane during $a$-plane GaN growth.\textsuperscript{27,28} It is important to note that these stacking faults propagate to the $a$-plane sample surface, and may therefore intersect any active area of a device grown on such layers. In fact, the high density of BSFs in the windows results from the propagation of the BSFs generated in the $a$-plane GaN template freely into the windows, while they are blocked by the SiO$_2$ mask on the wing regions. The existence of BSFs in the wing regions could be partially due to the misalignment between the growth windows and the GaN $m$-axis, as will be discussed below.

Dislocations were also found at the meeting fronts of sample A, as in the case of $c$-plane ELO.\textsuperscript{29} As evidenced in Figure 16(b), sample B also displayed nearly two orders of magnitude reduction of dislocations in the wings and generation of new dislocations at the meeting fronts. When the neighboring wings coalesce, a grain boundary is formed on the prismatic plane with the shift vector parallel to [0001]. These boundaries sometimes eliminate BSFs propagating from the substrate, but often are the sources of new defects propagating to the surface.

![Figure 15. (2110) Kikuchi lines from large angle convergent beam electron diffraction at the meeting front of the two opposing wings for sample A. Note that the Kikuchi lines are shifted across the meeting front.](image)
2.2e Atomic force microscopy (AFM) images

Tapping-mode AFM measurements were conducted to study the surface morphology of fully coalesced sample B. As shown in Figure 17(a), the surface exhibits striated features along the $c$-axis in both the window and the wing regions. Contrary to the $c$-plane GaN ELO, the meeting fronts are higher than the windows by about 60-100 nm. This is due to a 1.05° miscut of the $r$-plane sapphire towards its [0001] $c$-axis determined from XRD analysis. As illustrated in Figure 13(c), this miscut results in the inclination of the GaN $a$-plane towards [0001] by 1.05°, which is larger than the wing-tilt of ~0.4°, and an upward slope of the N-polar wings with respect to the substrate surface, giving rise to the elevation at the meeting fronts after coalescence. This inclination of the GaN $a$-plane, which is congruent with the miscut of the $r$-plane sapphire substrate, has its origin in the dominance of the GaN growth along its $c$-axis. This is the direction along which the lateral growth fronts in $a$-plane ELO advance. With increasing substrate miscut, the GaN $a$-plane and the wings are still expected to follow the miscut angle.
Growth on $m$-plane sapphire (that may be considered as 32.4° miscut of the $r$-plane away from the $c$-axis) has been shown to produce $a$-plane GaN surfaces inclined by 32.4° with respect to the substrate. This important observation may pave the way for epitaxial growth of patterned GaN surfaces with well-defined crystal planes (both polar and nonpolar) on miscut sapphire substrates without the need for additional processing steps.

AFM measurements on the as-grown $a$-plane GaN ELO samples revealed significantly different densities of surface pits in the window and wing regions, which are usually attributed to dislocation terminations at the surface. Figure 17(b) shows an AFM image near the window and N-polar wing boundary for sample B, where the pit density in the windows ($\approx 3.0 \times 10^9$ cm$^{-2}$) is approximately two orders of magnitude larger than that in the wings ($\approx 3.7 \times 10^7$ cm$^{-2}$). The pit density deduced from AFM is smaller than the dislocation density obtained from TEM, most probably because pure edge dislocations are harder to observe in AFM, making the apparent total dislocation density smaller. However, both AFM and TEM results confirm the effective dislocation reduction in $a$-plane by ELO. Additionally, an etch pit density study was also carried out to estimate the dislocation densities. As shown in Figure 18(a), the pits in the windows were enlarged and became more obvious after etching in a mixed $\text{H}_3\text{PO}_4/\text{H}_2\text{SO}_4$ (1:1) solution for 20 min at 200 °C, and the pit densities were similar to those before etching. In the wing regions, the etching has generated clear but inhomogeneously distributed steps extending along the GaN $m$-direction. In the N-polar wing regions, the steps were uniform and dense, while in the Ga-polar wing regions, most steps were observed near the windows, and the sectors near the meeting fronts were free of steps. The distribution of these steps is very similar to that of the stacking faults observed from cross-sectional TEM in Figure 18(b). The density of these etching-formed steps is around $2 \times 10^4$ cm$^{-1}$ in the Ga-polar wings and $6 \times 10^4$ cm$^{-1}$ in the N-polar wings.
respectively, giving an average density of $3-4 \times 10^4 \text{ cm}^{-1}$ in the wings. These values are in good agreement with those obtained from TEM. Currently, there is no report regarding wet etching of non-polar GaN and the origin of these steps. Wet chemical etching has often been employed in the study of bulk SiC growth to reveal stacking faults on cleavage planes, and BSFs have been observed as linear steps on wet-etched prismatic planes of SiC.\textsuperscript{32,33,34} Therefore, we suggest that the steps formed after etching of the $a$-GaN ELO samples might be due to the presence of BSFs propagating to the surface. Therefore, wet-etching offers a very convenient and effective means to characterize the structural defects, especially the stacking faults in non-polar or semi-polar GaN.

As shown in Figure 18(a), there is a small misalignment (around 2-4°) between the windows and the steps (oriented along the GaN $m$-axis) formed after etching that represent the BSFs propagating to the surface. This indicates that the windows, i.e. the SiO$_2$ stripes are not perfectly aligned along the $m$-axis of GaN, which may have led to the observation of BSFs in the wing areas near the windows. Extreme caution should be taken during the alignment of the SiO$_2$ mask for non-polar ELO as BSFs could propagate from the template and through the windows into the wings if the windows are not oriented parallel to the GaN basal plane.\textsuperscript{35}
Figure 17. (a) 30 µm × 30 µm AFM image for sample B. (b) 4 µm × 4 µm AFM image near the window and N-polar wing boundary of sample B, showing different surface pit densities for the window and the wing.
Figure 18. (a) 20 µm × 20 µm AFM image for sample B after etching in a mixed H₃PO₄/H₂SO₄ (1:1) solution for 20 min at 200 ºC. (b) Cross-sectional TEM image for sample B showing the distributions of basal stacking faults in the wings, with most of them located near window regions.
2.2f Near-field scanning optical microscopy (NSOM)

In order to assess the improvement in optical quality with the two-step ELO, NSOM was carried out on samples B and C. Figure 19(a) and (b) show the AFM and NSOM images, respectively, taken from a 40 µm x 40 µm area of sample B. The window and wing regions, as well as the meeting fronts are clearly distinguishable from the NSOM image of Figure 19(b). The windows appear as dark regions, and the narrow and the wide bright regions are due to N- and Ga-polar wings, respectively, with no significant difference between the intensities from the two. The reduced PL intensity at the meeting fronts may be partly due to a probe-size-related artifact as a weak drop has also been observed in the near-field reflection intensity (reflection of 325 nm laser light) at the meeting fronts. The larger size of the NSOM probe (350 nm aperture size) compared to conventional AFM tips may result in the partial blockage of the luminescence or the reduction of the excitation density when it experiences abrupt height changes related to surface features. The increased PL intensity in the wing regions of the ELO samples compared to the window regions suggests improvement of the material quality in the wings by reduction of dislocations (also verified by TEM measurements discussed above) that act as and/or cause nonradiative recombination channels. Similar to sample B, sample C (20 µm stripe sample) also showed a weaker average PL intensity in the window regions and at the meeting fronts compared to the wing regions [see Figure 19(c) and (d)]. However, the N-wings in sample C as compared to the Ga-wings demonstrate a weaker PL intensity, suggesting inferior optical quality of the N-wings. Additionally, the average PL intensity from the Ga-wings in sample C is less than one sixth of that in sample B. These results indicate that the material quality is certainly inferior for sample C, which also exhibited a larger (11̅20) reflection FWHM value of 0.27º for φ=0º.
2.2g Photoluminescence (PL) results

Low temperature PL spectra were measured for sample B using 325 nm excitation from a HeCd laser, as shown in Figure 20. The near bandedge emission at 15 K is composed of two peaks at 3.475 and 3.419 eV [see Figure 20(a)]. The 3.475 eV peak, which has a FWHM value of 19 meV, is a combination of the free exciton and donor bound exciton transitions, which are broad and cannot be delineated even at 15 K. As shown in Figure 20(b), this main peak slightly
blueshifts with increasing temperature up to 50 K and then starts to redshift reaching 3.415 eV at 300 K. This blueshift up to 50 K is due to the dissociation of the bound exciton with increasing thermal energy, which makes the free exciton peak dominant. The peak at 3.419 eV for the 15 K PL is most probably due to recombination of carriers/excitons bound to stacking faults, and also to other structural defects on the surface. The band edge emission from sample B is approximately two orders of magnitude stronger than that from an a-plane GaN template on r-plane sapphire without ELO under identical measurement conditions [see Figure 20(a)]. The characteristic blue and yellow emission bands are also observed with peaks around 2.96 and 2.25 eV, respectively. The blue band is possibly associated with the transitions from the conduction band or shallow donors to Zn-related acceptors. As shown in Figure 20(a), the blue band and the band edge luminescence weaken significantly at 300 K while the intensity of the yellow band remains nearly unchanged.

Time-resolved PL measurements were also performed for sample B at room temperature. Figure 21 shows the 200 μJ/cm² excitation, bandedge TRPL data (normalized to one) for the a-plane GaN ELO sample B and also for a standard a-plane GaN template. The decays for all the samples can be well characterized by a biexponential decay function $A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2)$. The fitting parameters for sample B are $\tau_1=0.08$ ns, $\tau_2=0.25$ ns, and $A_2/A_1=1.63$ at the bandedge emission peak energy. Both of the measured decay times, $\tau_1$ (short) and $\tau_2$ (long), are limited by nonradiative recombination, and longer decay times and larger $A_2/A_1$ ratios indicate reduced nonradiative relaxation pathways. The inset of Figure 21 shows the room-temperature near bandedge PL spectrum along with the effective PL decay times $\tau_{eff} = (A_1\tau_1 + A_2\tau_2)/(A_1 + A_2)$ measured at different photon energies. The representative $\tau_{eff}$ near
the band edge is approximately 0.2 ns. The radiative ($\tau_R$) and nonradiative ($\tau_{NR}$) lifetimes can be roughly estimated from the effective PL decay times using $1/\tau_{eff} = 1/\tau_R + 1/\tau_{NR}$ and substituting the equivalent internal quantum efficiency $\eta = 1/(1 + \tau_R / \tau_{NR})$ with the spectrally integrated PL intensity at room temperature normalized to that at 10 K ($\eta = 0.032$ at room temperature). The resulting $\tau_R$ and $\tau_{NR}$ values are 6.45 and 0.21 ns, respectively. The decay times for the $a$-plane ELO sample are significantly longer compared to the standard $a$-plane GaN template (see Figure 21), for which the system response (~40 ps) was not fast enough to resolve the exact PL decay. The improvement in decay times with $a$-plane ELO suggests reduction of nonradiative channels caused by the structural defects and the associated point defects, consistent with the TEM and NSOM observations discussed above.

Figure 20. (a) PL spectra for sample B at 15 K and 300 K and for the control $a$-plane GaN sample at 15 K. (b) PL spectra for sample B at temperatures 15, 25, 50, 75, 100, 150, 200, 250, and 300 K (from top to bottom and vertically shifted for clarity).
Figure 21. Normalized room temperature time-resolved PL for Sample B and a standard $a$-plane GaN template. Solid line represents the biexponential fit to data. The inset shows the near bandedge PL at room temperature and the effective PL decay times ($\tau_{\text{eff}}$) at different photon energies.

2.2h Conclusions

By employing a two-stage ELO of $a$-plane GaN with an initially enhanced vertical growth followed by enhanced lateral growth at an elevated temperature, Ga to N wing width ratio, and therefore, the wing height difference at the coalescence fronts have been reduced, resulting in a relatively flat, fully coalesced surface. TEM studies indicated the formation of new boundaries on inclined prismatic planes and dislocations along the vertical growth direction at the coalescence fronts due to the formation of a step between the two wings. In spite of this, the threading dislocation density was reduced from $4.2 \times 10^{10}$ cm$^{-2}$ in the window regions to $1.0 \times 10^{8}$ cm$^{-2}$ in the wing regions, which still had a relatively high basal stacking fault density of $1.2 \times 10^{4}$
cm$^{-1}$. The improvement in the overgrown layer quality by ELO was also verified by NSOM and TRPL measurements, with the former showing strongly enhanced luminescence from the wing regions, while the latter indicated longer decay times compared to a standard $a$-plane GaN template.

2.3 Epitaxial lateral overgrowth of $(1 \bar{1}00) m$-plane GaN on $m$-plane 6H-SiC

Growth of $(1 \bar{1}00) m$-plane GaN has been receiving considerable attention to alleviate the spontaneous and strain-induced piezoelectric polarization effects that are inherent to the $c$-plane hexagonal GaN system. Investigations about $m$-plane GaN growth have not been very extensive. So far, only a few reports on growth of $m$-plane GaN on $\gamma$-LiAlO$_2$ (100)$^8$ and $m$-plane SiC substrates$^9$ exist. In order to reduce the defect density, epitaxial lateral overgrowth has been carried out using hydride vapor phase epitaxy$^{40}$. In this section, we report the growth and characterization of ELO $m$-plane GaN on an $m$-plane 6H-SiC substrate using MOCVD.

2.3a Experimental procedures

The $m$-plane GaN films investigated in this study were grown on $(1 \bar{1}00) m$-plane 6H-SiC substrates [less than 0.1° miscut as measured by XRD] using MOCVD. Prior to growth, SiC substrates were annealed ex-situ in H$_2$ atmosphere at 1500 °C for 10 min to remove the polishing-induced surface damage. Trimethylgallium and ammonia were used as the Ga and N sources, respectively. Following in-situ annealing of the chemically cleaned SiC substrate, a 100 nm-thick high-temperature AlN nucleation layer was deposited at 1050 °C. The temperature was then ramped down to 1030 °C for the subsequent growth of the $m$-plane GaN epilayer. The ELO
of $m$-plane GaN was carried out using a mask with periodic stripes. The growth mask was an 80 nm-thick SiN$_x$ layer deposited using plasma-enhanced chemical vapor deposition. A mask pattern with 10 $\mu$m-wide stripes and 4 $\mu$m-wide windows was transferred onto the template using standard lithographic procedures, with the stripes oriented along the [11\overline{2}0] $a$-axis of the $m$-GaN template. The flow rates of TMGa and NH$_3$ were kept at 117 $\mu$mol/min and 550 sccm, respectively, with the growth pressure kept at 30 Torr.

2.3b Results and discussion

Figure 22 shows the XRD data for a planar $m$-plane GaN sample grown on a 6H-SiC substrate by MOCVD. The growth plane was determined to be the GaN $\{1 \overline{1} 00\}$ $m$-plane using a symmetric 2$\theta$-$\omega$ scan (not shown here), which revealed only a GaN (10 $\overline{1}$0) and a (20$\overline{2}$0) $m$-plane peak together with a (10 $\overline{1}$0) $m$-plane AlN peak. In order to determine the in-plane epitaxial relationship between the GaN and the SiC substrate, off-axis diffraction scans were performed by changing the $\phi$ and $\psi$ angles, where $\phi$ is the angle of rotation about the sample surface normal and $\psi$ is the angle of tilt about the axis formed by the intersection of the Bragg and scattering planes. According to the results shown in Figure 22, the in-plane relationship between $m$-GaN and $m$-SiC is given below: $[11\overline{2}0]_{GaN} \parallel [11\overline{2}0]_{SiC}$ and $[1 \overline{1} 00]_{GaN} \parallel [1 \overline{1} 00]_{SiC}$. Surface morphology revealed by SEM [see Figure 23(a)] and AFM [see Figure 23(b)] showed striated features, which are very similar to those of a typical (11$\overline{2}$0) $a$-plane GaN epilayer\textsuperscript{41}. However, the orientation of the striations of $m$-plane GaN was along the GaN [11$\overline{2}$0] direction, as determined by XRD, while that for $a$-plane GaN was along the GaN [0001] direction. The root-mean-square (RMS) roughness for this $m$-plane GaN template over a 5 x 5 $\mu$m$^2$ area was 2.8 nm.
Figure 22. Off-axis XRD scans to determine the epitaxial relationships between $m$-plane GaN and $m$-plane 6H-SiC.

Figure 23. (a) A typical SEM image of an $m$-plane GaN template grown on an $m$-plane SiC substrate. The directions were determined by XRD, (b) A tapping-mode AFM image (5 x 5 $\mu$m$^2$) of an $m$-plane GaN template, with a RMS roughness of 2.8 nm.
In order to reduce the extended defect density, ELO was also carried out on an $m$-plane GaN template with the stripe orientation along the GaN $[1\bar{1}20]$ $a$-axis. The schematic is shown in Figure 24(d). This stripe direction is expected to reduce the stacking fault density in the wings, based on our results from the ELO of $(1\bar{1}20)$ GaN$^{42}$ and also $m$-plane GaN ELO by the HVPE method$^{43}$. After 1 h of regrowth, the sample was taken out from the MOCVD chamber for SEM observation. As shown in Figure 24(a), the sidewall of the Ga-face ([0001]) wing was composed of one crystalline facet, while the N-face wing has a vertical sidewall. The facet of the Ga-face wing makes an angle of around 28º with the GaN $m$-plane, according to a cross-sectional SEM image (not shown), which suggests it to be the GaN $(1\bar{1}01)$. After 2.5 h of regrowth, the opposing wings began to meet each other [see Figure 24(b)], and the wings coalesced after a cumulative 4.5 h of regrowth [see Figure 24(c)]. However, this coalescence is not uniform: uneven trenches could still be observed in some areas of this sample, as shown in the inset of Figure 24(c). These trenches generally disappeared slowly with further growth, while some of them persisted during growth, even at a much higher growth temperature (1070 ºC). This could be due to the relatively good stability of the GaN $(1\bar{1}01)$ plane during MOCVD growth$^{44,45}$ and also the lower lateral growth rate of the N-face wings.
Figure 24. Plan-view SEM images for ELO sample with (a) 1h regrowth, (b) 2.5h regrowth, (c) 4.5h regrowth, respectively. (d) The schematic for the $m$-plane GaN ELO process. The inset in (c) shows a typical trench found in some areas of the sample.
Tapping-mode AFM images from the overgrown area of the ELO m-GaN sample show clear atomic steps [see Figure 25], which are different from the striated features observed on the planar m-GaN template. The absence of the striated features on the ELO sample could be indicative of the reduction of basal stacking faults in the ELO wing regions. XRD ω-scans show significant improvement of the crystalline quality. The FWHM values of the on-axis XRD rocking curves for the GaN (1 ̅00) after ELO were 580 and 2490 arcsec when rocked toward the GaN a-axis and c-axis, respectively, compared to 940 and 4570 arcsec from the m-GaN template. The FWHM values of off-axis XRD rocking curves are 1180 arcsec for GaN (1 1 ̅20) when rocked toward the GaN c-axis, and 1070 arcsec for GaN(1 ̅01) when rocked toward the GaN a-axis.

Figure 25. A tapping-mode AFM image (4 x 4 μm²) of m-plane GaN ELO from the overgrown area (∆Z=10 nm).

PL measurements were carried out on the ELO sample to characterize the optical quality of the thin films. As shown in Figure 26, the room-temperature PL is dominated by a near-band edge emission at 3.41 eV and a broad yellow luminescence at around 2.2 eV. However, at 10K
the near bandedge emission is composed of two peaks at 3.47 and 3.41 eV (see Figure 26). The 3.47 eV peak is due to the main donor bound exciton transition, while the peak at 3.41 eV for the 10 K PL is most probably due to recombination of carriers/excitons bound to basal stacking faults (BSFs)\textsuperscript{47}, considering the fact that the PL data were collected not only from the wings but also from the windows, which contain a large number of BSFs. The peak at 3.29 eV and the small broad peak at 3.34 eV are possibly associated with dislocations terminating at the BSFs and with prismatic stacking faults, respectively\textsuperscript{47}. Additionally, at 10K the characteristic blue and yellow emission bands are also observed with broad peaks centered at 2.95 and 2.25 eV, respectively.

![Figure 26. PL spectra for an $m$-plane GaN ELO sample at 10K and 300K, with the inset showing the details of the near-band edge emission.](image)

Figure 26. PL spectra for an $m$-plane GaN ELO sample at 10K and 300K, with the inset showing the details of the near-band edge emission.
2.3c Conclusions

An $m$-plane GaN film has been grown on an $m$-plane 6H-SiC substrate and the epitaxial relationship between the $m$-GaN and the substrate was analyzed. The ELO process results in a significant improvement of the crystalline quality in $m$-GaN according to AFM and XRD measurements. Low-temperature PL spectra show an exitonic transition at 3.47 eV, BSF-related emission at 3.41 eV, and other defect-related emission lines at 3.29 eV and 3.34 eV.

2.4 Epitaxial growth of m-plane GaN on Si(112) substrates

The advantages of using silicon substrates for the growth of GaN include the availability of large-size wafers with high crystalline quality at reasonably low cost, good thermal conductivity, and also ease with which they can be selectively removed before packaging for better light extraction and heat transfer when needed. Contrary to the growth of $c$-plane GaN on Si(111), which has been explored extensively with considerable success, the studies of non-polar or semi-polar GaN growth on Si have been limited. The epitaxial relationship between hexagonal GaN(0001) and Si(111) substrates has been well understood: GaN$\langle0001\rangle ||$Si$\langle111\rangle$ and GaN$\langle2\bar{1}0\rangle ||$Si$\langle011\rangle$. By patterning Si wafer to create Si(111) planes, the growth of (1120) $a$-plane non-polar GaN on patterned Si(110), (1122) semi-polar GaN on patterned Si(113), and (1T01) semi-polar GaN on patterned Si(001) has been achieved; however, studies of $m$-plane nitrides and related LEDs on Si substrates have not yet been reported. According to the epitaxial relationship between GaN and Si, the growth of GaN on Si(112) should result in an $m$-plane orientation (i.e., GaN $m$-plane parallel to the sample surface). In this case, growth must be initiated on Si(111) facets, which requires patterning of the Si...
substrate to expose the \{111\} facets and allow only \{\overline{1}1\} facets to participate in growth. In this work, we describe the epitaxial growth of non-polar, \textit{m}-plane GaN on stripe-patterned Si(112) substrates using MOCVD.

\textbf{2.4a Experimental procedures}

Si(112) substrates were patterned prior to growth to expose vertical Si\{111\} facets as shown in Figure 27. First, a 100 nm-thick SiN\textsubscript{x} layer was deposited by plasma-enhanced chemical vapor deposition on a Si(112) substrate\textsuperscript{56}, and was then patterned to form a wet-etch mask having periodic stripes (4 \textmu m-wide mask plus 10 \textmu m-wide open window) along the Si[\overline{1}1\text{0}] direction by using inductively-coupled plasma etching after standard photolithography. Subsequent anisotropic wet-etching of Si was performed in a KOH solution (4.16 Molar) at 40 °C to form Si\{111\} facets. As shown in Figure 27, after KOH etching two groups of Si\{111\} planes would be formed, one being perpendicular to Si(112) and the other forming an angle of 19º with the Si(112) surface. The vertical Si\{\overline{1}1\} sidewalls are used for the \textit{m}-GaN growth and have a depth of approximately 4 \textmu m. Even though the depth could be increased by increasing the period of the SiN\textsubscript{x} etching mask, this was deemed unnecessary because full coalescence of the subsequently grown GaN film would then require longer growth times.

Next, a 100 nm-thick AlN layer was grown by MOCVD on the patterned Si substrate at 1050 °C to serve as a seed layer for the GaN growth after the remaining SiN\textsubscript{x} etching mask was removed using buffered-oxide etch. To initiate growth only on the vertical Si\{\overline{1}1\} sidewalls and not on the Si(112) and inclined Si\{111\} surfaces, a SiO\textsubscript{2} mask was then deposited using angled e-beam evaporation to expose only the vertical Si\{\overline{1}1\} sidewalls [Figure 28]. The Si substrate was then reloaded into the MOCVD chamber for GaN growth, which was initiated on the vertical
Si$(\overline{1}1\overline{1})$ planes. Growth advanced laterally first along the GaN [0001] $c^+$ direction and then additionally along the [000$\overline{1}$] $c^-$ direction after the vertical growth advanced above the Si(112) substrate plane, resulting in a coalesced $m$-plane GaN surface after a sufficient amount of growth. For the GaN growth, during the first 30min, a growth temperature of 1000°C, a chamber pressure of 76 torr and an ammonia flow rate of 2L were used, while during the rest 2.5h period, a growth temperature of 1030°C, a chamber pressure of 30 torr and an ammonia flow rate of 550sccm were used. Trimethylgallium flow rate was kept at 117 $\mu$mol/min.

![Diagram](image)

Figure 27. Schematic depiction of selective area $m$-plane GaN growth on patterned Si(112) substrates. Since growth is initiated on the vertical Si$(\overline{1}1\overline{1})$ sidewalls, the threading dislocations propagate along the c-axis and not toward the surface.
2.4b Results and discussion

Figure 29 shows the plan-view and cross-sectional scanning electron microscopy (SEM) images of the m-plane GaN after 3 hours of growth. As seen from the figure, the GaN layer was partially coalesced [see Figure 29(a)] because of an insufficient overgrowth time and also in part because of a relatively low ratio of lateral growth rate to vertical growth rate (1:1 in this case) under the non-optimized GaN growth conditions employed. From the cross-sectional SEM images in Figure 29(b) and Figure 29(c), the GaN growth was confirmed to initiate only on the vertical Si(111) sidewalls. With further growth and coalescence, the facets and trenches observed in Figure 29(c) at the meeting fronts are expected to gradually disappear and result in a smooth m-plane GaN surface. Figure 30 shows a tapping-mode atomic force microscopy (AFM) image of the m-plane GaN sample on Si. The image indicates a very smooth surface (RMS roughness of ~0.3 nm over an area of 2 µm × 2 µm) with clear atomic steps, suggesting a step-flow growth mode for this sample.
Figure 29. (a) Plan-view and (b) angled cross-sectional SEM images of an $m$-GaN sample grown on a patterned Si(112) substrate after a 3 h growth. (c) Enlarged cross-sectional SEM image showing the growth of the $m$-GaN initiated at the vertical Si(111) sidewalls.

Figure 30. Tapping-mode AFM image ($\Delta z = 10$ nm) of an $m$-plane GaN sample (~12µm thick) grown on a patterned Si substrate. The AFM image indicates a very smooth surface (RMS roughness of ~0.3 nm over an area of 2 µm$\times$2 µm) with clear atomic steps.
On-axis XRD $2\theta$-$\omega$ scans were performed to confirm the GaN orientation, with the results shown in Figure 31(a). The XRD $2\theta$-$\omega$ scan data only revealed GaN $m$-plane (1 $\bar{1}$00) and (2200) diffraction peaks, while the peak from the Si(112) substrate does not appear due to a diffraction extinction effect. To determine the in-plane epitaxial relationship between the GaN and Si substrate, off-axis XRD scans were performed by changing the $\phi$ and $\psi$ angles, where $\phi$ is the angle of rotation about the sample surface normal and $\psi$ is the angle of tilt about the axis formed by the intersection of the Bragg and scattering planes. As shown in the inset of Figure 31(a), GaN(1 $\bar{1}$01) is oriented 180° away from the tilted Si(111) in terms of $\phi$ angles which conforms to the relationship depicted in the schematic of Figure 27. To determine the crystalline quality of the grown $m$-plane GaN sample XRD rocking curves were measured. As shown in Figure 31(b), the rocking curve full width at half maximum (FWHM) is 9 arcmin when rocked toward the GaN $a$-axis, and 27 arcmin when rocked toward the GaN $c$-axis. Because the GaN $c$-axis, which is perpendicular to the stripes, is the laterally advancing direction for the $m$-GaN growth, the larger FWHM value in the case of rocking toward the $c$-axis reflects broadening due to tilting of the advancing wings in the $c^+$ and $c^-$ directions. However, the individual XRD linewidths of the wings were most probably not sufficiently narrow to allow the determination of the tilt angle.
Figure 31. (a). On-axis XRD ω-2θ scan showing that the GaN has (1100) m-plane orientation. The Si(112) plane does not have a diffraction peak due to diffraction extinction. The inset shows the off-axis scan results, suggesting that GaN(1T01) is oriented 180° away from Si(111). (b) XRD rocking curves of GaN(1T00) m-plane after a 3 h growth, rocking toward the GaN a-axis and c axis.

It should be mentioned that cracking of GaN on Si(111) is a major problem due to the inherent tensile strain induced by the very large difference of thermal expansion coefficient between GaN (Δa/a: 5.59×10⁻⁶/K, Δc/c: 3.17×10⁻⁶/K) and Si (2.6×10⁻⁶/K) unless special strain-releasing layers are employed. Because growth in our case is done on patterned Si substrates, such cracking has been mostly avoided. Only a very limited number of cracks were observed on the sample surface along the GaN c-axis and were separated by hundreds of microns, because the GaN stripes are contiguous along the GaN a-direction. Using low-temperature AlN⁵⁷,⁵⁸ or AlN/GaN superlattices⁵⁹ as a buffer layer, the cracking in our case may be avoided. In order to characterize the optical properties of the m-plane GaN on Si, photoluminescence (PL) measurements were performed using a HeCd laser operating at 325 nm wavelength, and the recorded spectra revealed that the bandedge emission of m-GaN on Si was approximately 75% of that measured for a c-plane epitaxially lateral overgrown (ELO) GaN sample on c-plane sapphire of comparable thickness. This is remarkable considering that the m-plane GaN layer is on a Si
substrate and the ELO growth of c-plane GaN have undergone years of improvement. For further evaluation of the optical quality, 6-nm thick InGaN double heterostructure (DH) light emitting diode active layers were deposited by MOCVD both on the m-GaN sample on Si and on a high quality c-GaN template grown on c-sapphire. The InGaN layers were capped by 100-nm p-type GaN. A frequency-doubled femtosecond Ti:sapphire laser emitting at 370nm was used for excitation dependent PL measurements from which the internal quantum efficiencies (IQEs) were deduced using the method described in Ref. 60, but with Auger effects included. The IQE measurement results of the InGaN layers on Si have been reported previously. 61 The InGaN PL emission peak blueshifts with increasing excitation density for the sample on c-plane substrate [Figure 32(a)], while no shift is observed for the InGaN DH structure on m-GaN [Figure 32(b)], indicating the absence of polarization fields. When compared to its c-plane counterpart, the IQE of the InGaN DH layer on m-GaN on Si is twice as high [see Figure 32(c)] and reaches 65% at a steady state carrier density of 1.2×10^{18} \text{ cm}^{-3}, corresponding to the maximum excitation density employed and the radiative recombination coefficient assumed (B = 10^{-11} \text{ cm}^3\text{s}^{-1}).
Figure 32. Room-temperature PL spectra measured with a He-Cd laser at different excitation densities for 6 nm-thick InGaN double heterostructure LED active layers on (a) c-GaN on sapphire and (b) m-GaN on Si. No blueshift is observed in the emission peak of the m-plane sample. The excitation power densities in (a) and (b) were 0.05, 0.15, 0.52, 1.0, 2.0, 2.5, 5.1 kW/cm$^2$. Figure (c) shows the internal quantum efficiencies of both samples extracted from the excitation dependence of the PL intensity using a Ti-sapphire laser (370nm).

In order to obtain a spatial distribution of luminescence properties of the InGaN sample on Si(112), cathode-luminescence (CL) measurements were carried out on the sample at low temperature (5K). Figure 33(a) shows the SEM image for the region where the CL measurements were carried out. The spatially integrated CL spectrum is shown in Figure 33(d). In addition to the emission from InGaN at 386 nm, well-defined peaks corresponding to excitons/carriers bound to basal stacking faults (BSFs) and prismatic stacking faults (PSFs) are visible in the spatially integrated CL spectrum at 363 nm and 373 nm, respectively, which are also
characteristic for a-plane GaN.\textsuperscript{62} This emission line has been also observed in 10-K PL spectra of an m-plane ELO GaN layers grown on m-plane SiC substrates.\textsuperscript{63} Furthermore, in order to identify the distribution of the above-mentioned emission lines, the spatially resolved CL image was spectrally integrated only within a 2 nm bandwidth for the BSF peaks and 12 nm bandwidth for the InGaN peak. As shown in Figure 33(b), the emission at 363nm attributed to BSFs in m-plane GaN\textsuperscript{63} is relatively stronger for the nitrogen-polar wings [see Figure 33(a)], suggesting that the N-polar wings of the laterally overgrown areas have significantly higher density of BSFs than the overgrown Ga-polar wings. A similar BSFs distribution has been observed in laterally overgrown a-plane samples.\textsuperscript{64} Moreover, N-polar wings (which show stronger BSF-related emission) exhibits relatively weaker InGaN emission [see Figure 33(c)]. The PSF related emission (CL image not shown) was found to be nearly uniform across the sample where InGaN was deposited and it showed essentially the same spatial distribution as that of the emission from InGaN. This indicates that PSFs are uniformly distributed across the InGaN layer, as V-defects characteristic to InGaN promotes formation of additional defects such as the PSFs.\textsuperscript{48}

To complement the CL measurement of defects in the m-GaN on Si, microstructure of the GaN grown on patterned Si (112) substrates was studied using transmission electron microscopy (TEM). Figure 34 shows a bright-field TEM image of the N-polar wing. In an agreement with the CL results, multiple BSFs are clearly seen as dark lines and dashes perpendicular to the [000\overline{1}] direction in the micrograph. The dimensions of staking faults on the basal plane vary from a few nanometers to several micrometers. The density of threading dislocations is low in the N-wing, which might be the reason for the clearly observed BSF-related emission in the N-polar wings in CL, in the light of a recent report of anti-correlation between the light emission from radiative centers, including the BSF line, and the density of
dislocations in non-polar GaN films.\textsuperscript{65} It is also worth noting that some areas of the material are virtually free of the extended defects (not shown).

Figure 33. (a) SEM image of the m-plane InGaN sample on Si(112), (b) Spatial distribution of integrated CL intensity with wavelength from 362nm to 364 nm, which is attributed to BSF-related emission, (c) Spatial distribution of integrated CL intensity with wavelength from 384nm to 396nm, which is attributed to InGaN emission. Brighter regions in (b) and (c) indicate higher intensity. The scale bar for images (a), (b) and (c) is shown in (a). (d) Spatially integrated CL spectrum from the image shown in (a). The results suggest that BSFs are only located in the area where the vertical Si(111) sidewalls and the sample surface are intercepted.
2.4c Conclusions

The epitaxial growth of $m$-plane GaN films has been achieved on the vertical Si$(\overline{1}1\overline{1})$ sidewalls of patterned Si(112) substrates, as confirmed by on-axis XRD measurements. Remarkably, internal quantum efficiencies of InGaN DH layers grown on these $m$-GaN films on Si are twice that of $c$-plane varieties, reaching 65% at a moderate carrier density of $1.2 \times 10^{18}$ cm$^{-3}$. BSFs-related CL emission is only observed in the overgrown N-polar wings, while the CL spectra from Ga-polar wings are dominated by InGaN emission. TEM shows a low density of threading dislocations in the N-polar wings. The selective area growth of non-polar, $m$-plane GaN layers on patterned and relatively low cost Si substrates is very promising for high efficiency light emitters.
2.5 Epitaxial lateral overgrowth of (11\overline{2}2) semi-polar GaN on \textit{m}-plane sapphire

Employing semi-polar GaN is another option to reduce polarization effects. (10\overline{1}3) or (11\overline{2}2) oriented semi-polar GaN has been grown on \textit{m}-plane sapphire using hydride vapor phase epitaxy (HVPE),\textsuperscript{9} and electroluminescence from semi-polar InGaN/GaN light emitting diodes has showed a reduced blue shift with increasing drive current compared to the \textit{c}-plane counterparts, indicative of reduced polarization in the active layer.\textsuperscript{10} However, no systematic study of semi-polar GaN growth by MOCVD on nominal \textit{m}-plane sapphire has been reported. Next, we describe a detailed investigation of GaN growth on \textit{m}-plane sapphire using MOCVD, which could also provide better understanding of GaN growth on non-polar sapphire substrates, and ultimately help to realize high material quality in this orientation.

2.5a Experimental procedures

GaN films investigated in this study were grown on nominally on-axis (10\overline{1}0) \textit{m}-plane sapphire substrates [less than 0.1° miscut as measured by XRD] using MOCVD. Trimethylgallium and ammonia were used as the Ga and N sources, respectively. Following \textit{in situ} annealing of the chemically cleaned sapphire substrate, a 45 nm-thick low-temperature GaN nucleation layer was deposited at 550 °C. The temperature was then raised to 1030 °C for the subsequent growth of a 1.2 \(\mu\text{m}\)-thick GaN epilayer. The ELO of GaN on \textit{m}-plane sapphire was also carried out using (11\overline{2}2) GaN films as the template. The growth mask was a 140 nm-thick SiO\textsubscript{2} layer deposited using plasma-enhanced chemical vapor deposition. A mask pattern with 10 \(\mu\text{m}\)-wide stripes and 4 \(\mu\text{m}\)-wide windows was transferred onto two different pieces of the
template using standard lithographic procedures: one having stripes oriented along the [1210] $a$-axis and the other along the [0001] $c$-axis of the sapphire substrate. The two samples were grown side by side under identical conditions in the MOCVD chamber. The flow rates of TMGa and NH$_3$ were kept at 117 $\mu$mol/min and 550 sccm, respectively, with the growth pressure at 30 Torr. The growth time was kept short (1 h) and coalescence of the wings was avoided so that advancement of the growth fronts could be observed.

2.5b Results and discussion

To determine the crystallographic orientation and the crystalline quality, the as-grown GaN films on $m$-plane sapphire were characterized by high-resolution XRD. As shown in Figure 35(a), on-axis $2\theta$-$\omega$ scans indicate that the surface orientation is $(11\overline{2}2)_{\text{GaN}} \parallel (10\overline{1}0)_{\text{sapphire}}$. The full width at half maximum (FWHM) values of XRD rocking curves for the on-axis GaN$(11\overline{2}2)$ and off-axis GaN$(0002)$ shown in Table 1 for different rocking directions are similar to those reported for GaN films grown on $m$-sapphire using HVPE.\textsuperscript{9} [see also Table 1]. However, unlike the case of HVPE, no $(10\overline{1}3)$ oriented GaN films were obtained even after high-temperature nitridation of the sapphire substrate in NH$_3$ (550 sccm) atmosphere at 1030 °C for 2 min or an increase of NH$_3$ flow rate (from 550 sccm to 7060 sccm) during the initial stages of the epitaxial growth.

| Table 1. FWHM values (in arcsec) of XRD rocking curves for $(11\overline{2}2)$ GaN templates, ELO samples with mask stripes along the $a$-axis and $c$-axis of sapphire, and an $a$-plane GaN ELO sample for different rocking directions. |
In order to obtain the in-plane epitaxial relationship, off-axis $\phi$ scans were performed at different $\psi$ tilt angles for the (1122) GaN film. From the pole figure in Figure 35(a) inset, it is concluded that $(0002)_{\text{GaN}}$ is oriented $180^\circ$ away from $(01\overline{1}2)_{\text{sapphire}}$. The GaN (1122) plane forms an angle of $58.4^\circ$ with $(0002)_{\text{GaN}}$, while the sapphire $(01\overline{1}2)$ plane forms an angle of $32.4^\circ$ with the sapphire $(10\overline{1}0)$ $m$-plane. After combining these results, one determines the epitaxial relationships for GaN on $m$-plane sapphire, as shown in Figure 35(b); namely,

$$(11\overline{2}2)_{\text{GaN}} \parallel (10\overline{1}0)_{\text{sapphire}}, [10\overline{1}0]_{\text{Ga}N} \parallel [\overline{1}2\overline{1}0]_{\text{sapphire}}, [1\overline{2}1\overline{1}]_{\text{GaN}} \parallel [0001]_{\text{sapphire}}.$$

Figure 36(a) shows a plan-view scanning electron microscopy (SEM) image of the GaN (1122) film grown using 550 sccm NH$_3$ flow rate as described above. The image is indicative of a rough surface with “V”-shaped features (with pits placed at the corners) that are oriented along the $c$-direction of the sapphire substrate. Although the orientation of the GaN films was not sensitive to the growth conditions in this study, the surface morphology of the samples exhibited slightly different features under different growth conditions, especially with changing ammonia flow rate. Based on the growth conditions for the sample shown in Figure
36(a), when the NH₃ flow rate was changed from 550 sccm to 7060 sccm for the first 10 min of the epilayer growth, different surface features were observed, as shown in Figure 36(b). The pits in this image are surrounded by four facets, two of which make an angle of approximately 110º, which is similar to the V-shaped features in Figure 36(a), while the other two make an angle of approximately 50º. The facets for the V-shaped features or pits seem to be the GaN \{10\bar{1}1\} planes, which are very stable and often observed during MOCVD growth of c-plane and a-plane GaN. The angle between the two neighboring \{10\bar{1}1\} planes is 127.6º which would be observed as 113º or 50º when projected onto the GaN (11\bar{2}2) surface, consistent with the figures measured from the SEM images in Figure 36. The observed angle values, however, show a wide distribution between 90º and 115º, most probably due to varying amount of deposition on these planes. The structure of the V-shaped feature is clearly seen in the 3D atomic force microscopy (AFM) image of Figure 36(c).
Figure 35. (a) XRD $2\theta$-$\omega$ scan for GaN grown on $m$-plane sapphire, with the inset showing the off-axis $\phi$ scans with different $\psi$ tilt angles (i.e., pole figure) for GaN (0002) and sapphire (0112) of GaN(1122) on $m$-plane sapphire. (b) A schematic depicting the epitaxial relationship derived from XRD measurements for (1122) GaN on $m$-plane sapphire.
Figure 36. Plan view SEM images of GaN samples grown on \( m \)-plane sapphire using an NH\(_3\) flow rate of (a) 550 sccm, (b) 7060 sccm for the first 10 min and 550 sccm for the rest of the growth. The arrows indicate the \( c \)-axis of sapphire. (c) A 3D AFM image (3 x 3 \( \mu \)m\(^2\)) of a V-shaped surface feature (\( \Delta z = 150 \) nm).

In order to reduce the extended defect density and also to study the lateral growth behavior of this semi-polar film, ELO was carried out on a (11\( \bar{2}2 \)) GaN template (grown using 550 sccm NH\(_3\)) with the stripe orientations described above. Figure 37(a) shows a plan-view SEM image for the ELO sample with SiO\(_2\) stripes oriented along the \( a \)-axis of the sapphire substrate. It is clear from the cross-sectional SEM image in Figure 37(b) that the wings for this mask orientation are inclined by 32° with respect to the substrate plane with well-defined \( a \)- and \( c \)-plane surfaces. The growth along the \( c \)-axis advances faster than that in other directions; therefore, the observation of inclined wings is consistent with the epitaxial relationships shown in Figure 35, which suggest a 32.4° angle between the \( c \)-axis of GaN and \( c \)-axis of sapphire. The upwardly inclined wings are tentatively attributed to the Ga-polar (0001) wings, since their
growth fronts (GaN c-plane) are smooth with rare occurrences of some facets as observed in the SEM images, while the N-polar (000 T) surfaces often feature hexagonal hillocks.\textsuperscript{12} Because of the large incline angle, only the Ga-polar wings extend while the N-polar wing growth is stymied by the template. With further growth on this ELO sample, the Ga-polar wings extended further along the c-axis of GaN with negligible growth along the a-axis. Unlike the GaN(1122) surface of the template shown in Figure 36, both a- and c-plane surfaces of the wings are very smooth. AFM measurements on the a-plane surfaces of the wings revealed no striated features along the c-axis that are characteristic of the a-plane GaN growth on r-plane sapphire,\textsuperscript{4} and a root-mean-square surface roughness of ~ 2 nm, which is much smaller than that of the fully coalesced a-plane GaN ELO layers (9-15 nm).\textsuperscript{13}

Compared to the (1122) GaN templates on m-plane sapphire and also a-plane GaN ELO samples,\textsuperscript{13} significant improvement of the crystalline quality has been achieved with ELO, as exemplified by the XRD rocking curve data summarized in Table 1. The FWHM values of the XRD rocking curves for the on-axis GaN (1122) after ELO reduced to 380 and 610 arcsec when rocked toward the GaN m-axis and c-axis, respectively. This suggests significant reduction in the defect density,\textsuperscript{14} as confirmed by the cross-sectional transmission electron microscopy (TEM) image shown in Figure 37(c). The high number of BSFs that appear in the template and also in the window regions are effectively eliminated in the overgrown Ga-wings, which exhibit an average dislocation density of only ~10\textsuperscript{7} cm\textsuperscript{-2}.  

66
Figure 37. (a) Plan-view, (b) cross-sectional SEM, and (c) cross-sectional TEM images of GaN ELO sample with SiO₂ stripes oriented along the \( a \)-axis of sapphire. (d) Plan-view, (e) cross-sectional SEM, and (f) cross-sectional TEM images of the GaN ELO sample with SiO₂ stripes oriented along the \( c \)-axis of sapphire.

For the second ELO sample, the SiO₂ mask stripes were oriented along the \( c \)-axis of the sapphire substrate, which is also along GaN \([1\overline{2}1\overline{1}]\). In this stripe orientation, the lateral overgrowth should progress along the \( m \)-axis of GaN. Figure 37(d) shows the plan-view SEM
image for this ELO sample. The cross-sectional SEM image in Figure 37(e) indicates the formation of two different planes that make an angle of 26° with the substrate plane. Using XRD studies these surfaces were identified as \{10\overline{1}1\}. According to the XRD rocking curve results shown in Table 1, no improvement of the crystal quality is achieved for this stripe orientation when compared to the (11\overline{2}2) GaN templates on \textit{m}-plane sapphire and the ELO sample with SiO\textsubscript{2} stripes oriented along the \textit{a}-axis of the sapphire. From the cross-sectional TEM image of this sample [see Figure 37(f)], it is evident that the BSFs (observed as horizontal lines which represent the cross section of these planar defects with the [1\overline{2}1 \overline{1}] plane of GaN) cannot be blocked but propagate laterally in the overgrown material for this particular stripe orientation. In addition, the threading dislocations originating from the template bend towards the two \{10\overline{1}1\} surfaces and are partially eliminated only in a very limited part of the overgrown volume. Therefore, both XRD and TEM results suggest that no significant dislocation density reduction occurs when the stripes in the mask are oriented along the \textit{c}-axis of sapphire.

\textbf{2.5c Conclusions}

In summary, epitaxial growth of GaN on \textit{m}-plane sapphire substrate was carried out on \textit{on-axis} (10\overline{1}0) \textit{m}-plane sapphire using MOCVD. XRD measurements revealed a uniquely preferred (11\overline{2}2) orientation of the GaN films. For the ELO samples with stripes aligned along the \textit{a}-axis of the sapphire substrate (lateral growth along the \textit{c}-direction), SEM images showed 32.4° inclined Ga-polar wings, with well-defined GaN \textit{a}-plane and \textit{c}-plane surfaces. When the stripes were aligned along the \textit{c}-axis of the sapphire (lateral growth along the GaN \textit{m}-direction), the surface was composed of two GaN \{10\overline{1}1\} planes each making a 26° with the substrate plane.
Significant improvement of the crystalline quality was achieved by ELO with relatively small amount of overgrowth only when the mask stripes were oriented along the sapphire $a$-axis according to XRD rocking curve and TEM measurements. Moreover, AFM and XRD results also suggest that the extended nonpolar GaN $a$-plane with mask stripes along the sapphire $a$-axis has better surface morphology and crystalline quality than that of $a$-plane GaN ELO, which would suggest a promising route for the growth of high-quality nonpolar GaN.
Chapter 3. LED performance on non-polar GaN substrates

3.1 Introduction

In order to clarify any correlation between efficiency droop and polarization-induced electric field and or the extent of it, we have undertaken an investigation of the m-plane light emitting diodes (LEDs) and their electroluminescence (EL) efficiency degradation behavior. In addition to the lack of polarization induced field, the non-polar m-plane GaN, on which InGaN/GaN LEDs material is grown, has been predicted to have a few advantages such as a reduced hole effective mass, shallower acceptor level and thus higher hole density. It is well known that, in the c-oriented GaN films, the internal spontaneous and strain-induced piezoelectric polarizations produce a strong electric field. This results in the spatial separation of electron and hole wavefunctions in quantum wells (QWs) used in LEDs and laser diodes (LDs), thereby reducing the quantum efficiency, particularly at low injection levels.\textsuperscript{66,67} The polarization-induced electric field also causes a red shift in LEDs and makes the emission wavelength dependent on injection current, blue shifting with increasing injection, unless very thin quantum wells are employed. By employing non-polar orientations, namely m-plane\textsuperscript{8,68} or a-plane GaN,\textsuperscript{69,70} this problem could be circumvented. As noted above, m-plane GaN is purported to be well-suited for optoelectronic applications in that it is predicted to have a reduced valence band effective mass (thus smaller acceptor binding energy), and larger optical matrix elements relative to its c-plane counterpart, which would bode well for LEDs and LDs on this orientation.\textsuperscript{71} The abovementioned reduced hole effective mass would also help to obtain a relatively high hole density.\textsuperscript{72,73} This in turn would increase hole injection into the recombination medium and thus increase the radiative recombination rate as well as increasing the hole density.
in wells farther away from the p-n junction. In this regard, the use of non-polar orientations would assuage carrier spillover and help in the efficiency retention at high injection levels.

3.2 Experimental procedures

All LED structures were grown on freestanding GaN in a vertical low-pressure metalorganic chemical vapor deposition (MOCVD) system. They are composed of 6 period 2 nm In$_{0.14}$Ga$_{0.86}$N quantum wells with 12 nm In$_{0.01}$Ga$_{0.99}$N barriers, and a 60 nm Si-doped (2×10$^{18}$ cm$^{-3}$) In$_{0.01}$Ga$_{0.99}$N underlayer just beneath the active region for improved quality. A ~10 nm $p$-Al$_{0.15}$Ga$_{0.85}$N electron blocking layer was deposited on top of the active quantum well region. The Mg-doped $p$-GaN layer that followed is about 100 nm thick having 7×10$^{17}$ cm$^{-3}$ hole density for the c-plane variety, as determined by Hall measurements on a calibration sample, which is expected to be higher for the m-plane orientation due to lighter hole effective mass for the same Mg chemical content. Due to the fact that the m-plane LED sample is extremely small, we did not perform Hall measurements. Further details and the schematic of the LED structures can be found in Ref. 74. After mesa (250 μm diameter) etching, Ti/Al/Ni/Au (30/100/40/50 nm) metallization annealed at 800 °C for 60 seconds was used for $n$-type ohmic contacts and 5 nm/5 nm Ni/Au contacts were used for the semi-transparent $p$-contacts. Finally, 30/50 nm Ni/Au contact pads were deposited on the top of part of the mesa. The 500 μm-thick $m$-plane freestanding GaN templates, produced at Kyma Technologies, Inc., have a threading dislocation density of <5x10$^6$ cm$^{-2}$ and are off-cut by 0.2° towards the GaN $a$-axis and 0.3° towards the GaN $c$-axis. The $c$-plane freestanding GaN is around 250 μm thick.

The internal quantum efficiency (IQE) values were extracted from the excitation-dependent PL studies with resonant excitation at room temperature using a frequency-doubled 80 MHz repetition rate femtosecond Ti:Sapphire laser. The excitation laser wavelength was 370 nm
whose energy is below the bandgap of the quantum barriers and top GaN, but higher than that of
the wells. In this scenario, the photo-excited electron-hole pairs are generated only within the
quantum wells where they are forced to recombine either radiatively or nonradiatively. In terms
of some details of efficiency determination, we have used a procedure similar to that described in
section 1.5 of chapter 1 to measure the IQE with the power-dependent PL.

3.3 m-plane LED vs. c-plane LED

The IQE values extracted from the excitation-dependent PL method for m-plane and c-
plane InGaN layers are shown in Figure 38. During the calculation of the carrier densities for the
samples under investigation, as noted above, the B coefficient was assumed to be $1 \times 10^{-11} \text{cm}^3\text{s}^{-1}$. At relatively high carrier densities, the IQE values of the $m$-plane LEDs are $\sim$30 % higher than those of their $c$-plane counterparts measured in our laboratory in one set of experiments. At the highest excitation power, the IQE values are 80 % and 60 % for the m-plane and c-plane LEDs, respectively.

In terms of the electroluminescence (EL) performance, the m-plane LED shows higher
EL intensity and better efficiency retention with increasing current injection. The on-wafer EL
measurements were carried out for both m-plane and c-plane LED samples without any special
treatment to enhance light extraction and efforts were undertaken to assure the same photon
collection geometry from sample to sample during measurements. The light vs. current
characteristics were measured under pulsed injection current (1 $\mu$s, 0.1 % duty cycle) in order to
minimize, if not totally eliminate, the heating effect on LED efficiency.
Figure 38. Extracted IQE values for the m- and c-plane LEDs on bulk GaN substrates using excitation intensity dependent PL measurements. The two LEDs have the same structure: 6 period 2 nm In$_{0.14}$Ga$_{0.86}$N quantum wells with 12 nm In$_{0.01}$Ga$_{0.99}$N barriers. During the calculation of carrier densities for the samples, the B coefficient was assumed to be $1 \times 10^{-11}$ cm$^3$s$^{-1}$.

Figure 39 shows the integrated EL intensity and relative external quantum efficiency (EQE) as a function of the injection current density for the two LEDs investigated. The m-plane LED shows negligible efficiency loss, i.e. almost full retention of its efficiency up to 2500 Acm$^{-2}$, which represents the highest current density under which the experiments were conducted. Quantitatively speaking, only 5% efficiency loss with the increase of current for the LED on m-plane as compared to ~25% for that on c-plane freestanding GaN having the same structure was noted.

The above observation is consistent with the premise of relatively high hole density in m-plane that would provide higher hole densities in the recombination region and reduce the electron spillover (or overflow) and thereby mitigate the efficiency loss. Furthermore, as also evident from Figure 39, at relatively low injection levels the EL intensity for the m-plane LED increases more rapidly than that for the c-plane LED, reaching its peak value at ~140 Acm$^{-2}$.
compared to ~400 Acm\(^{-2}\) for the c-plane LED, which is indicative of a relatively small Shockley-Reed-Hall nonradiative recombination coefficient for the m-plane variety. Among several devices tested for both orientations, the m-plane LED EQE values are consistently higher by ~35\%. This difference is even higher at higher injection levels due to better efficiency retention than those of the c-plane LEDs which is consistent with the IQE results obtained from the intensity dependent PL measurements. The variation from device to device for each orientation was less than 10\%.

Figure 39. (a) Integrated EL intensity and (b) relative external quantum efficiency of the m-plane LED on freestanding GaN and the reference LED on c-plane bulk GaN as a function of pulsed injection current density (1 \(\mu\)s, 0.1 \% duty cycle). Both samples have the same device structure (MQW active region with 2 nm In\(_{0.14}\)Ga\(_{0.86}\)N quantum wells, 12 nm In\(_{0.01}\)Ga\(_{0.99}\)N barriers, and p-Al\(_{0.15}\)Ga\(_{0.85}\)N electron blocking layers).
3.4 Polarized electroluminescence (EL) spectra from m-plane LEDs

The m-plane LEDs exhibit polarized EL due to the in-plane polarization anisotropy in m-plane, which is further enhanced by large valence band splitting induced by the anisotropic biaxial strain within the quantum wells.\textsuperscript{75,76} Three m-plane LEDs were investigated in this section, and they have the same structure as described in the experimental part, with different emission wavelengths (400nm, 435nm and 485nm, respectively) which were obtained by varying the growth temperature of the active region (see Figure 40 for the EL emission from the LEDs). Figure 41(b) shows the EL intensity of an m-plane LED as a function of the polarization analyzer angle, where 0º corresponds to polarization perpendicular to the c-axis. The measurement was done in a configuration as shown in Figure 41(a). The EL light was collected by a spectrometer which was positioned above the LED chips. As can be seen the electric field component of the EL is mainly polarized in the GaN m-plane and perpendicular to the GaN c-axis. The polarization ratio is given by \( \rho = (I_{\perp c} - I_{|| c}) / (I_{\perp c} + I_{|| c}) \), where \( I_{\perp c} \) and \( I_{|| c} \) correspond to intensities for polarization perpendicular and parallel to the c-axis, respectively. The polarization ratio values are calculated to be \(~0.49\), \(~0.60\), and \(~0.8\) for the 400 nm, 435 nm and 485nm emission wavelength m-plane LEDs, respectively. The degree of polarization increases with increasing emission wavelength. This phenomenon is consistent with what has been observed by others,\textsuperscript{77} and is attributed to enhanced valence-band splitting caused by larger compressive strain in QWs with increasing In composition.\textsuperscript{78}
Figure 40. m-plane non-polar LEDs emitting at (a) 400nm (near ultraviolet), (b) 440nm (blue) and (c) 490nm (blue-green) at DC current ~100 Acm$^{-2}$. The LEDs were grown on nonpolar m-plane GaN substrates.
Figure 41. Normalized EL intensity as a function of the polarization analyzer angle for $m$-plane LEDs (6 period 2 nm InGaN QWs and 3 nm InGaN barriers) with different peak emission wavelengths. The polarizer angles of 0° and 90° correspond to $E\perp c$ and $E\parallel c$, respectively.
Chapter 4. Efficiency droop in polar and non-polar InGaN LEDs

4.1 What is efficiency droop

Even though the performance of the state-of-the-art InGaN light emitting diodes (LEDs) has improved substantially over the decades, the InGaN-based LEDs suffer from loss of efficiency at high injection levels needed, which has been dubbed as the “efficiency droop”.\textsuperscript{79} The LED external quantum efficiency (EQE) reaches its peak value at relatively low current densities (10–50 A/cm\textsuperscript{2}) and monotonically decreases with further increase of current (see Figure 42 for an example of a commercial InGaN LED).\textsuperscript{4} The “droop” issue is observed even under pulsed current condition, which has basically eliminated the junction heating effect on the EQE.

Figure 42. EQE and integrated EL intensity of a typical InGaN LED under pulsed current condition (1\% duty cycle, 1kHz). The EQE reaches its peak value at relatively low current density of 50Acm\textsuperscript{2}, and then monotonically decreases with the increase of current density. At the current of 1400Acm\textsuperscript{2}, the EQE drops to 60\% of its peak value. In some LEDs, the magnitude of drop is even larger. (from a commercial company, $\lambda$=460nm)
4.2 What causes efficiency droop

Despite substantial progress on the performance front, the physical origin of the efficiency drop in InGaN LEDs is not understood with sufficient clarity as of yet and is open to further investigations, a prerequisite for the remedies to be found and implemented. Among the various causes proposed for the observed efficiency degradation are Auger recombination, current roll-off resulting from phonon-assisted tunneling through defect sites, poor hole injection, polarization field, and junction heating, as well as current crowding and related contact degradation. Junction heating can be mitigated by increasing the efficiency and employment of packages capable of removing the dissipated heat very efficiently, both of which are the subject of vigorous developmental efforts in industry. For the purpose of determining any role of junction heating pulsed measurements with low duty cycle are typically employed.

Despite being considered rather consistently by some as the origin of efficiency degradation in InGaN LEDs, the intrinsic Auger losses in wide bandgap semiconductors involving just the conduction band and the valence band are expected to be relatively small, as verified by fully microscopic many body models, but there are some new nuances which will be discussed below. The available data in aggregate appear to suggest that the Auger recombination alone (if any) might not be dominant as the observed efficiency droop depends on structural details of the active region, beyond the electron density.

Complicating the matters or clarifying them some depending on one’s point of view further is the absence of the efficiency loss in photo excitation experiments where carriers are photo-generated directly and only in the active region even with generation rates orders of magnitude larger than those encountered in high electrical injection levels. This instead suggests that the efficiency loss at high current is related to carrier injection processes (that is
electron overflow or spillover) rather than Auger recombination. In the following sections, we will give a brief description about Auger recombination process and show why Auger recombination is not the major reason for the efficiency loss at high current. We propose the electron overflow or spillover to be the dominant mechanism responsible for the efficiency loss issue. The term “spillover electrons” refers to the electrons which escape the active region without participating in any recombination process, radiative and non-radiative alike, and end up recombining in the p-GaN region or make it to the p-contact if the minority carrier lifetime in that region permits it. We will present a theoretical model to describe the electron overflow process, and will also propose a possible solution (using staircase electron injector) to eliminate the electron overflow.

4.3 Auger recombination

Let us now discuss the processes commonly attributed to causing efficiency degradation. Carrier loss due to Auger recombination, which is nonradiative in nature, has been proposed early on as the origin of the efficiency drop.\textsuperscript{80,93} Auger recombination is a three-carrier process such as CCCH (conduction band\to conduction band and conduction band\to heavy-hole valence band transitions), which depicts the process in which an electron gains the energy given off by another electron dropping to the valence band. Naturally both the energy and momentum must be conserved and as one can surmise the energy given off when dropping to the valence band does not lead to a radiative process. The aforementioned one together with those depicted by nomenclatures CHHS (conduction band\to heavy-hole valence band and spin-orbit split-off band\to heavy-hole valence band transitions) and CHHL (conduction band\to heavy-hole valence band and light-hole band\to heavy-hole valence band transitions) represent the intrinsic Auger processes. There are also other intrinsic processes which include phonons and extrinsic varieties.
involving defects. To reiterate, the CCCH process could be visualized as scattering involving two electrons, with one being knocked down from conduction band to the valence band and the other being scattered to a higher energy state in the conduction band. The latter would eventually thermalize down to the bottom of the conduction band, releasing the excess energy as heat to the crystal lattice. The CHHS and CHHL processes could be viewed as collisions of two holes within the heavy hole band, with one excited to the conduction band and the other sent deeper to either the split-off valence band or the light hole valence band, all obeying the momentum and energy conservation. Since the Auger processes depend on carrier collisions, Auger recombination rate highly depends on the carrier density, and thus gains more relevance at high injection current levels for optoelectronic devices. It also strongly depends on the bandgap energy ($E_g$) and temperature ($T$). For a semiconductor with a conduction band effective mass smaller than the valence band effective mass, the lifetime is determined by electron-electron collisions, namely electron recombination with a hole followed by another electron excitation to a higher energy while the entire process having to conserve the energy and momentum.

The net Auger recombination rate can be expressed as:

$$U_{Auger} = C_n \left[ n \left( p n - n_i^2 \right) \right] + C_p \left[ p \left( p n - n_i^2 \right) \right]$$

**Equation 5**

where $C_n$ and $C_p$ represent the Auger recombination coefficients for electrons and holes, respectively, $p$ and $n$ are the hole and electron densities given by $p = p_0 + \Delta p$, $n = n_0 + \Delta n$, $n_i$ is the intrinsic carrier density. Here $\Delta p$ and $\Delta n$ represent the excess hole and electron densities which are equal to each other when equal injection takes place such as in optical excitation. The first term in Equation 5 is due to the CCCH Auger processes and the second term is due to the CHHS process. This equation is valid only under non-degenerate conditions since Boltzmann
approximation is employed for the derivation of the equation. For degenerate semiconductors, Equation 5 overestimates the Auger recombination rate. Since GaN is nominally $n$-type ($p_0 << n_0$) and at high injection $Δp >> n_0$, $Δp >> p_0$, the above expression could be simplified to

$$U_{\text{Auger}} = C_n p n^2 + C_p p^2 n$$

Equation 6

In the case where $Δn = Δp >> n_0$, the electron and hole densities can be assumed equal leading to the Auger rate being $C_n n^3 + C_p n^3$, which represents a third power dependence on the carrier densities.

The Auger coefficient deduced through a rate equation fit to photoluminescence (PL) data in an earlier effort is $1.4-2.0 \times 10^{-30}$ cm$^6$/s for quasi-bulk InGaN layers. A recent calculation based on the first-principle density-functional and many-body theory reported the Auger recombination coefficient to be as large as $2 \times 10^{-30}$ cm$^6$/s. It must be noted, however, that this coefficient is said to result from the upper conduction band which would be in resonance for LEDs emitting at 2.5 eV. In this case, the Auger coefficient due to the aforementioned resonance should drop off precipitously on either side of the corresponding wavelength. In the absence of this resonance, the direct Auger recombination coefficient decreases exponentially with increasing bandgap energy and the carrier loss due to Auger effect is expected to be very small in InGaN if we were to use the size of the forbidden gap alone. Further credence to a small Auger coefficient is provided by fully microscopic many body models. If inherent and intrinsic Auger recombination were to be solely responsible for the efficiency degradation, one would surmise that it would have prevented laser action in InGaN, which requires high injection levels, which is not the case. The vexing phonon assisted and extrinsic Auger recombination, however, could be examined to determine whether this phenomenon could be in play here. To make matters more
complex, through fitting the EL efficiency data from commercial LEDs to the rate equation, which includes Auger recombination in addition to the Shockley-Read-Hall (SRH) recombination and bimolecular radiative recombination, Ryu et al.\textsuperscript{96} found unreasonably large Auger recombination coefficients in the range of $10^{-27}$-$10^{-24}$ cm$^6$s$^{-1}$ when the non-radiative lifetime $\tau_{SRH}$ is assumed to be between 5-50 ns. The deduced Auger coefficients are at least three orders of magnitude larger, if not more, than the other reported values which are also extracted from the efficiency dependence on injection.\textsuperscript{80,95,97,98} The wide variation, in fact some 6 orders of magnitude, might signal that the Auger recombination alone if any is not sufficient to explain the efficiency drop in InGaN LEDs. Furthermore, the extraction of the Auger coefficient assumes that the A, B, and C coefficients used to calculate the carrier loss rate in conjunction with the following equation

$$\frac{dn}{dt} = An + Bn^2 + Cn^3$$

\textbf{Equation 7}

do not depend on the electron density (injection levels) and also valid for any electron densities. The conditions under which Equation 7 would hold can be found in ref. 4. We should underscore that in below the barrier resonant photo excitation experiments (photons absorbed only in the quantum wells with ensuing generation of equal number of electrons and holes followed by either radiative or non radiative recombination only) the efficiency degradation has not been detected at carrier generation rates comparable to electrical injection where efficiency degradation has been observed which indicates that efficiency degradation might be related to the carrier injection, such as electron overflow.\textsuperscript{74,85}

The efficiency droop in LED EL data could not only be fitted by Auger recombination term, but also by using electron overflow term. To investigate whether the electron overflow
might be responsible for the efficiency droop as opposed to the Auger recombination, the recombination rate equation under steady state can be modified as \((J - J_{\text{spillover}}) / qd = An + Bn^2 + Cn^3 + J_{\text{spillover}} / qt\). When an empirical dependence of the spillover current on the injection current is chosen as \(J_{\text{spillover}} = k J^b\) with \(k\) and \(b\) being fitting parameters, and \(t = d [19] (n^3 \text{ like dependence if the radiative current dependence on electron density is used})\), reasonable fits to the experimental data from different LEDs can be obtained without the Auger term \((C = 0)\) but with electron overflow term.\(^99\) These results indicate that efficiency droop in blue LEDs can be empirically described by electron overflow without the need for Auger recombination.

4.4 Electron overflow

4.4a Evidence of electron overflow in InGaN LEDs

Experimentally the electron overflow has been shown to reduce the LED EL efficiency substantially. The electron overflow (or spillover) causes a substantial reduction (3-5 times) of the EL efficiency for LEDs without an electron blocking layer (EBL) in polar and m-plane non-polar orientations.\(^100\) In this section, we will present the EL results from LEDs with different EBL heights in both c-plane and m-plane orientations as an evidence of effect of electron overflow on the LED EL performance. Different EBL barrier heights have resulted in different amount of electron overflow and thereby different EL performance in LEDs.

The investigated m-plane LED structures were grown on freestanding m-plane (1\,Ì00) GaN substrates in a vertical low-pressure metalorganic chemical vapor deposition (MOCVD) system. The ~500 \(\mu\)m-thick m-plane freestanding GaN substrates, produced at Kyma
Technologies, Inc., had a threading dislocation density of $<5 \times 10^6 \text{ cm}^{-2}$ and were off-cut by 0.2° towards the GaN $a$-axis and 0.3° towards the GaN $c$-axis. The LED structures contain a 6nm undoped active region of $\text{In}_{0.20}\text{Ga}_{0.80}\text{N}$ ($\lambda_{\text{peak}} \approx 440\text{ nm}$) followed by a 3 nm undoped $\text{In}_{0.01}\text{Ga}_{0.99}\text{N}$ layer. Just beneath the 6 nm active layer, a 60 nm Si-doped ($2 \times 10^{18} \text{ cm}^{-3}$) $\text{In}_{0.01}\text{Ga}_{0.99}\text{N}$ underlayer was employed for improved active region quality unless otherwise specified. A $\sim 10$ nm EBL of $p$-$\text{Al}_x\text{Ga}_{1-x}\text{N}$ ($x=15\%$, or 8%, or 0%) was deposited on top of the active layer. The final Mg-doped $p$-$\text{GaN}$ layer was about 100 nm thick with a nominal hole density of $7 \times 10^{17} \text{ cm}^{-3}$.

The $c$-plane double-heterostructure (DH) LEDs investigated in the present work were grown on $c$-plane sapphire substrates with $\text{in-situ}$ SiN$_x$ nano-network, and have a 6nm $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}$ active region with a peak emission wavelength of $\sim 410\text{ nm}$. The structures of the $c$-plane LEDs are the same as their $m$-plane counterparts, except for the In composition (i.e. emission wavelength) of the active layer. After mesa ($250 \mu\text{m}$ diameter) formation, Ti/Al/Ni/Au (30/100/40/50 nm) metallization annealed at 800 °C for 60 seconds was used for $n$-type ohmic contacts, and 5 nm/5 nm Ni/Au semi-transparent p-contacts were employed. Finally, 50 $\mu\text{m}$ diameter 30/50 nm Ni/Au contact pads were deposited on part of the mesa tops for the on-wafer probed EL measurements.

The internal quantum efficiency (IQE) was extracted from excitation power-dependent PL measurements at room temperature using a frequency-doubled 80 MHz repetition rate femtosecond Ti:Sapphire laser. The excitation wavelength was 370 nm, below the bandgap of the barriers and the top GaN layer. On-wafer EL measurements were performed using pulsed current (0.1 % duty cycle, 1 kHz) without any special means to enhance light extraction.

According to our resonant PL measurements described above, the three LED structures have essentially the same IQE for the same injected carrier density (IQE of 50%-56% at a carrier
density of $1 \times 10^{18} \text{cm}^{-3}$ under the assumption that the radiative recombination coefficient $B$ is $1 \times 10^{-11} \text{cm}^3\text{s}^{-1}$). Thus, the LEDs with different EBLs demonstrate the same quality of the active layers without any systematic dependence on the EBL composition.

The results of EL measurement from the same LEDs are shown in Figure 43(a). The relative external quantum efficiency (EQE) as a function of the current density for the $m$-plane LEDs depends on the Al composition in their EBLs. The EQE for the $m$-plane LED with 15% Al in EBL (open squares in the figure) peaks at approximately 85 Acm$^{-2}$, and the efficiency drops by ~45% with increasing current injection up to 2400 Acm$^{-2}$. A negligible efficiency drop (~3-5%) is observed for the structure without an EBL (i.e., for 0% Al in EBL) with increasing injection. Another observation is that its relative EQE (Figure 43) is ~3-5 times lower than that with 15% Al in the EBL. Intermediate values for EQE and efficiency drop (~10%) are obtained for the LED with 8% Al within EBL (Figure 43). In general, the EQE decreases as the Al mole fraction in the EBL is reduced. As mentioned, the three investigated LED structures have essentially the same IQE obtained from PL measurements at the same photoexcited carrier density. It is, therefore, reasonable to suggest that the different EL efficiencies for LEDs with varying EBLs originate from the carrier injection and carrier overflow rather than the quality variation among the active regions.

If we assume that the LED with EBL having 15% Al has negligible electron overflow at a current density of 80 Acm$^{-2}$ and also the relatively low EL efficiency at current densities before reaching the peak efficiency for each LED is due to SRH recombination, we can then obtain the efficiency loss due to electron overflow according to the results in Figure 43(a). Figure 43(b) shows the percentile EL efficiency loss due to the electron overflow as a function of the applied voltage across the LEDs (including the voltage drop across the contacts). In the LED without the
EBL, 20% of the overall current contributes to the EL and approximately 80% of the current is due to electrons traversing the active region without contributing to the light emission. As we mentioned above, the voltage drop across the active region reduces the effective barrier by the p-side for the electrons, and therefore, results in increased electron overflow with increasing applied voltage. In the LED with 15% Al in the EBL, the efficiency loss at 16V is ~45%, which means that ~45% of the injected electrons are the overflow electrons since we have assumed, reasonably so, that this LED has negligible electron overflow at current densities at or below 80 Acm$^{-2}$. 
Figure 43. (a) Relative EQE of m-plane LEDs grown on freestanding m-plane (1 ̅100) GaN substrates with varying Al composition (15%, 8%, 0%) in the EBL layers, measured under pulsed current, 1 μs pulse width and 0.1 % duty cycle. The inset shows the current-voltage dependence for the LED with 15% Al in EBL. (b) The EL efficiency loss as a function of the external applied voltage across the p-n junction of the LEDs, assuming negligible electron overflow at the current density (~80 Acm⁻²) corresponding to the peak efficiency for the LED with 15% Al in EBL, which is very reasonable according to our calculations.

The c-plane LEDs contain polarization-induced electric field inside the c-plane InGaN active region and the EBL. The field has been proposed to be the reason for the electron...
overflow to the $p$-GaN layer. In this regard, we studied the $c$-plane LEDs as well to investigate the correlation between the polarization field and LED EL efficiency. The $c$-plane LEDs have a 6nm $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}$ as active region (DH LEDs) with an emission wavelength $\sim$410nm. Figure 44 shows the relative EQE data for two $c$-plane DH LEDs: one with a 10nm $\text{Al}_{0.15}\text{Ga}_{0.85}\text{N}$ EBL and one without any EBL. As is shown in the figure, the LED with EBL shows 3-4 times higher EL intensity than the one without any EBL. This indicates that as the case of $m$-plane LEDs, the absence of EBL has resulted in comparable amount of loss of the EL efficiency which is attributed to an increased electron overflow in the case without EBL. Thus the EBL can reduce the electron overflow in both $c$-plane and $m$-plane LEDs.

**Figure 44. Relative EQE of $c$-plane DH LEDs with and without the EBL layers. The LEDs were measured under pulsed current, 1 µs pulse width and 1 kHz frequency. The LEDs were grown on $c$-plane GaN templates on sapphire, and their active regions are composed of a 6 nm thick undoped $\text{In}_{0.15}\text{Ga}_{0.85}\text{N}$ layer (emission wavelength, $\lambda \sim$410nm).**

### 4.4b Electron overflow due to thermionic emission

At the onset with no *a-priori* inclination, we need to consider that the electron overflow might originate from two possible phenomena: ($i$) thermionic emission of thermalized injected
electrons from the bottom of the active region over the barrier into the p-layer (we will show this not to be important due to large band discontinuity for our investigated LEDs), (ii) ballistic and quasi-ballistic transport of the injected electrons that do not thermalize while in the active region. Let us consider the former point through numerical simulations using the Silvaco Atlas software for a p-GaN/In_{0.20}Ga_{0.80}N/n-GaN LED without an electron blocking layer (EBL). For the calculations, the electrons were assumed to be completely thermalized and in equilibrium in the active region. For this and also the rest of the calculations, the commonly accepted material parameters were used for the In_{0.20}Ga_{0.80}N active layer; a Shockley-Read-Hall (SRH) recombination coefficient (A) of 1×10^7 s^{-1}, a spontaneous radiative recombination coefficient (B) of 1×10^{-11} cm^3 s^{-1}, and an Auger recombination coefficient (C) of 1×10^{-30} cm^6 s^{-1} were assumed. For p-GaN, an A coefficient of 1×10^{10} s^{-1} (corresponding to a lifetime of 100 ps) was used. The conduction band offset ΔEc between In_{0.20}Ga_{0.80}N and p-GaN was deduced to be 0.5 eV by assuming 70% of the total band gap discontinuity.

The simulations involving the above-mentioned thermionic emission show that even at an uncharacteristically elevated junction temperature of 1000K and an unreasonably high current density of 1×10^4 Acm^{-2}, the thermionic emission driven overflow electron current into the p-GaN region is only ~11% of the total current density. At the same current density of 1×10^4 Acm^{-2}, the corresponding values are ~1% and ~0% for junction temperatures of 700K and 500 K, respectively. The calculated energy distributions of electrons for several junction temperatures are shown in Figure 45, where they are compared on a flat band diagram for simplicity. Keeping in mind the large discrepancy for the Auger coefficient,^{80,88,96} the calculated electron overflow values are 0%, 3% and 36% for junction temperatures of 500K, 700K and 1000K, respectively, if an Auger coefficient of 1×10^{-34} cm^6 s^{-1} is assumed. Notably, such a meager tail of the electron
distribution within a thermal energy (kT) of and above the barrier on the p-side of the junction cannot account for the observed 3-5 times EL difference between the LEDs with and without EBL as observed in section 4.4a. This indicates that the genesis of the notable electron overflow is not the thermionic emission of the equilibrium electrons from the active region even in the LEDs without EBL. We must therefore turn our attention to non-equilibrium electrons, in other words hot electrons, inside the In$_{0.20}$Ga$_{0.80}$N (the particular composition used in our experiments) active region. The injected hot electrons can traverse the active layer by ballistic or quasi-ballistic transport and recombine in the p-GaN region instead of the active region. We should note that the nonradiative recombination is prevalent in p-type GaN due to its defective nature, and any radiative recombination in the p-GaN would produce an unwanted wavelength in addition to that emanating from the designed active region.

![The overflow portion due to thermionic emission](image)

Figure 45. Calculated energy distribution of electrons in the active region at 3 different junction temperatures: 500K, 700K and 1000K. A flat band diagram schematic is used just to show the barrier height of the p-GaN and for a comparative picture of the three distributions, which actually correspond to three different band diagrams. The integrated tail of the Fermi-Dirac distribution
makes up only 0%, ~1% and ~11% of the total current at the junction temperatures of 500K, 700K and 1000K, respectively.

4.4c Hot electron overflow model

Having made the suggestion that the electron overflow current might be due to ballistic and quasi-ballistic transport of the injected electrons through the active region, in this section we will discuss a first-order estimation of the hot electron effect and attempt to explain our experimental data for LEDs with varying barrier height of the EBL. The calculation assumes that the electrons obey the Fermi-Dirac distribution in the n-GaN layer, i.e., before they are injected into the active region. The electrons acquire the additional kinetic energy equal to the conduction band offset between $n$-GaN and In$_{0.20}$Ga$_{0.80}$N ($\Delta E_c \sim 0.5\text{eV}$ in this case) upon injection. The kinetic energy means the additional electron velocity in the direction transverse to the $n$-GaN/In$_{0.20}$Ga$_{0.80}$N interface. These hot electrons would either lose their excess energy mainly through interaction with LO-phonons$^{103}$ and undergo thermalization, or avoid thermalization and leave the InGaN region as depicted in Figure 46.
Figure 46. Schematic of electron overflow caused by ballistic or quasi-ballistic electron transport across the InGaN active region. The electrons gain a kinetic energy after being injected into InGaN, which equals to $E + dE_c + qV(x)$. These hot electrons will either traverse the active region ballistically and quasi-ballistically, escape recombination inside InGaN, and contribute the electron overflow current, or be thermalized and captured inside the active region through interactions with LO-phonons.

Our calculations take into account the ballistic electrons, representing those that experience no scattering in the active region, and the quasi-ballistic electrons that experience one scattering event (i.e. quasi-ballistic motion involving either LO phonon emission or absorption), and two scattering events (4 combinations of two scattering events involving LO phonon emission and absorption). As will be evident soon those experiencing multiple energy loosing scattering events are eventually thermalized. In the calculations the electrons are categorized according to their scattering events that they experience: no scattering, one scattering event (one LO phonon emission or one LO phonon absorption), etc. We should mention that the calculated contribution of electrons undergoing two scattering events to the overflow, as we will demonstrate below, is less than 1% of the total injected electrons and can be ignored for a 6 nm thick active region as in our experimental device structures. Because of the negligible contribution of the electrons undergoing two scattering events, and necessarily beyond, to the
electron overflow, let us consider below only three cases: (i) – no scattering, (ii) – one phonon emission, (iii) – one phonon absorption.

1) **Case 1. Ballistic case, i.e. experiencing no LO phonon scattering**

As shown in Figure 46, at position \( x \) inside the InGaN layer, the kinetic energy of an electron is \( E + \Delta E_c + qV(x) \), where \( E \) is the excess energy in the n-GaN layer with respect to the conduction band of n-GaN, due to e.g. band bending and kinetic/thermal energy (assuming electrons have a Fermi-Dirac distribution in n-GaN), \( \Delta E_c \) is the conduction band offset between the InGaN and the n-GaN layers, \( V(x) \) is the net potential drop within the active region at position \( x \) due to the electric field (including the effect of the built-in potential) with boundary conditions of \( V(0)=0 \) and \( V(L)=V \), and \( L \) is the active region thickness. Therefore, the electron velocity at position \( x \) is \( v(E + \Delta E_c + qV(x)) = \sqrt{2\left[E + \Delta E_c + qV(x)\right]/m_e} \), where \( m_e \) is the electron effective mass. The total time for the electron to traverse the active region is then
\[
 t = \int_0^L dx / v[E + \Delta E_c + qV(x)] \]
. The probability of the ballistic process is given by: \( \exp\left(-t/\tau_{sc}\right) \), where \( \tau_{sc} \) is the phonon scattering time given by \( \tau_{sc} = 1/(1/\tau_{abs} + 1/\tau_{em}) \), where \( \tau_{abs} \) and \( \tau_{em} \) are the LO-phonon absorption and emission times, respectively. We should mention that the phonon scattering time is actually dependent on the electron density and energy, but we will assume it to be constant in the present treatment for simplicity.\(^{104} \)

The percentage of the overflow-electrons for case 1, which begin their journey in the n-GaN layer just prior to injection, is represented by the product of the probability of electrons having excess energy \( E \geq \max\left[0,(\phi_{eBL} - qV)\right] \) above the conduction band in the n-GaN layer and the probability of ballistic electron transport across the active region:
\[ P_i = \left[ \int_{\max [0, (\phi_{\text{EBL}} - qV)]}^{\infty} f(E) N(E) \exp \left( \int_0^E \frac{-dx/\tau_w}{v[E + \Delta E_T + qV(x)]} \right) dE \right] \left\{ \int_0^{\infty} f(E) N(E) dE \right\} \]

Equation 8

where \( f(E) \) is the Fermi-Dirac distribution function in the n-GaN, \( \phi_{\text{EBL}} \) is the barrier height of the EBL (i.e. the conduction band offset between the EBL material and the p-GaN, which is 0 for the case with no EBL), and \( N(E) \) is the conduction-band density of states in the InGaN layer.

The calculation of Equation 8 requires the potential distribution \( V(x) \) inside InGaN layer, which requires numerical simulation packages such as Silvaco-Atlas. Due to the lack of inclusion of the hot electron models in the packages, the effect of potential inside the active region (i.e. the acceleration or deceleration by the electric field) on the electron velocity is neglected at the moment for all cases in the present work. In the c-plane LED case this would underestimate the electron overflow percentile as we will show in section 4.4f. However, the barrier lowering at the p-region by the applied voltage across the active region has been taken into consideration in our model in order to account for the voltage dependent electron overflow. After neglecting the acceleration or deceleration of electrons by the electric field inside the active region, the exponential term in Equation 8 could be simply replaced by \( \exp \left[ -L/v(E + \Delta E_T)/\tau_w \right] \). For further simplicity, we assume that the electrons traverse only in the direction normal to the hetero-interface, which would insignificantly overestimate the probability of escaping scattering events and overestimate the current overflow some.

2) Case 2. One phonon emission during the electron transport across the well

Let us start from the known electron distribution \( f(E) \) in the n-GaN layer where the electrons are assumed to be in equilibrium with the lattice. Among the electrons, injected from
the n-GaN layer, only those which still have enough energy to surmount the barrier at the p-side after one phonon emission inside the active region will contribute to the overflow current. The total probability of an electron overflowing to the p-side via one phonon emission (LO phonon energy $\hbar\omega_{LO} = 88\text{meV}$) is the product of the following four partial probabilities:

(a) The probability of finding an electron in the n-GaN layer with an excess energy $E \geq (\phi_{inj} - qV + \hbar\omega_{LO})$ above the bottom of the conduction band of the n-GaN before injection into the active region:

$$P_a = \frac{\int_{E_{BL,LO}}^{\infty} f(E) N(E) dE}{\int_{0}^{\infty} f(E) N(E) dE}$$

**Equation 9**

(b) The probability to travel distance $x$ from the n-GaN/InGaN hetero-interface without being scattered:

$$P_b = \exp\left(-\frac{[x/v(E + \Delta E)]}{\tau_{em}}\right)$$

**Equation 10**

(c) The probability to emit an LO-phonon between $x$ and $x+dx$:

$$P_c = \left[\frac{dx/v(E + \Delta E)}{\tau_{em}}\right]$$

**Equation 11**

where $\tau_{em}$ is the phonon emission time.

(d) The probability to reach the opposite interface of the active region without being scattered:

$$P_d = \exp\left(-\frac{[(L-x)/v(E + \Delta E - \hbar\omega_{LO})]}{\tau_{em}}\right)$$

**Equation 12**

The overall probability or total percentile of the overflow electrons for one phonon emission, the case 2, is, therefore, given by:

$$P_2 = P_a \cdot P_b \cdot P_c \cdot P_d = \int_{E_{BL,LO}}^{\infty} f(E) N(E) \int_{0}^{L} \frac{\exp\left(-\frac{[x/v(E + \Delta E)]}{\tau_{em}}\right)}{v(E + \Delta E) \cdot \tau_{em}} dx \cdot \frac{dx/v(E + \Delta E)}{\tau_{em}} \cdot \frac{dx/v(E + \Delta E - \hbar\omega_{LO})}{\tau_{em}}$$

**Equation 13**

3) **Case 3. One phonon absorption during the electron transport across the well**
Similar procedures have been used as those of case 2, except for step (c), the phonon emission time was replaced with phonon absorption time, and the velocity after one absorption was \( v(E + \hbar \omega_{pb}) \). Moreover, the integration in the numerator was performed from \( E = \max\{0, (\phi_{\text{int}} - qV - h\omega_{pb})\} \) to \(+\infty\) to account for the electrons possibly involved in this single phonon absorption process. The total percentage of the overflow electrons for case 3 is therefore given by:

\[
P_3 = \int_{\max\{0, (\phi_{\text{int}} - qV - h\omega_{pb})\}}^{+\infty} f(E)N(E) \int_0^L \frac{\exp\left(-\left[\frac{x}{v(E + \Delta E_c)}\right]/\tau_w\right)}{v(E + \Delta E_c) \cdot \tau_{\text{abs}}} \times \exp\left(-\left[\frac{(L - x)}{v(E + \Delta E_c + h\omega_{\text{abs}})}\right]/\tau_w\right) \cdot dx \cdot dE \left(\int_0^\infty f(E)N(E)dE\right)
\]

*Equation 14*

Another possible source of electron overflow is due to the electrons that undergo two scattering events: (i) one phonon absorption followed by one phonon emission, (ii) one phonon emission followed by one phonon absorption, (iii) two phonon emission, (iv) two phonon absorption. For example, in a manner similar to that described above for single scattering events, the percentile of electron overflow for the case of “one phonon absorption followed by one phonon emission” can be written as:

\[
P_4 = \left(\int_0^\infty f(E)N(E)\,dE\right)^{-1} \int_{\max\{0, (\phi_{\text{int}} - qV - h\omega_{pb})\}}^{+\infty} f(E) \cdot N(E) \int_0^{x_1} \int_0^{x_2} \exp\left(-\left[\frac{x_1}{v(E + \Delta E_c)}\right]/\tau_w\right) \cdot \exp\left(-\left[\frac{(x_2 - x_1)}{v(E + \Delta E_c + h\omega_{\text{abs}})} + (L - x_2 - x_1)/v(E + \Delta E_c)\right]/\tau_w\right) \cdot v(E + \Delta E_c) \cdot \tau_{\text{abs}} \cdot \tau_{\text{em}} \cdot dE_1 \cdot dE_2 \cdot dE
\]

*Equation 15*

This equation takes into account the combined probabilities for an electron to reach the position \( x_1 \) without any scattering event, to absorb a phonon near \( x_1 \) (\( 0 \leq x_1 \leq x_2 \)), reach the position \( x_2 \) without being scattered, emit a phonon near \( x_2 \) (\( 0 \leq x_2 \leq L \)), and finally escape the active region without being scattered between points \( x_2 \) and \( L \). The integration over \( dx_1, \, dx_2, \) and
$dE$ accounts for all possible paths and all suitable electrons within the realm of processes discussed above. However, the contribution of these overflow-electrons undergoing two scattering events is negligible and accounts for less than 1% of the total electron overflow.

Figure 47(a) shows the calculated percentile electron overflow (i.e., overflow-electron current divided by the total current) as a function of $\phi_{EBL}$ for flat-band conditions in an $m$-plane LED which is void of any polarization-induced electric field. For all of the calculations, phonon emission ($\tau_{em}$), absorption ($\tau_{abs}$), and scattering times ($\tau_{sc}$) of 0.01ps, 0.1ps, 0.009ps, respectively, have been used.$^{104}$ An LO-phonon energy of $\hbar \omega_{LO} = 88$ meV has been used for InGaN layers.$^{105}$ To reiterate the aforementioned time constants are functions of the electron density, but in the present first order theory they will be assumed to be independent of electron density and energy. As mentioned, the calculated probabilities for the overflow by the electrons that undergo two or more scattering events are negligible ($\leq 1\%$). Under the flat-band conditions for the LED without EBL, meaning $y=0\%$ Al in Al$_y$Ga$_{1-y}$N EBL, a significant portion (~62%) of the total electron current is due to electrons escaping the active region altogether, while only 2% electron overflow occurs when an EBL with $y=15\%$ Al is inserted, as elaborated on below.

The calculated ratio of overflow-electron current to the total current is shown in Figure 47(b) as a function of the applied forward voltage ($V_T$) across the $m$-plane LEDs with different EBLs. The relationship between the internal voltage across the active region ($V$ in the above equations) and $V_T$ was obtained using Silvaco Atlas with simulation parameters given in section 4.4b, neglecting the voltage drop across the metal/semiconductor contacts. The large applied biases resulting across the devices despite the fact that voltage drops across the contacts were neglected, point at the limitations of the simulation package, which however represents a suitable tool to show the trends in electron overflow. According to the numerical simulations by Silvaco
Atlas, the flat band conditions in the active regions are achieved at $V_T = 3.8$, 4.0, and 4.7 V corresponding to $\sim 535$, 660, and 1150 A/cm$^2$ current injection for the LEDs with 15%, 8%, and 0% Al in the EBL, respectively. As shown in Figure 47, the calculated overflow current increases with the applied voltage, and the amount of electron overflow increases with decreasing EBL barrier height. For $V_T = 6$V, the calculated overflow current percentiles are 68%, 16%, and 2% for the LEDs with 0%Al, 8%Al and 15% Al in EBLs, respectively. Moreover, and not surprisingly, the percentile overflow for each LED shows a dependence on the applied voltage on the diode: when the voltage increases from 4V to 16V, the overflow increases from 56% to 80% for the LED with 0% Al in EBL, and from 0% to 42% for that with 15% Al in EBL.

It should be reiterated that this calculation is a simplified first-order estimation of the electron overflow; we assumed a constant phonon scattering rate regardless of the current. However, as has been reported\textsuperscript{104} the phonon lifetime decreases with increasing electron density, which translates to increasing current or the forward bias voltage, causing a decrease in the calculated electron overflow. In other words, our calculations would overestimate the dependence of electron overflow on applied voltage (or current). Secondly, the effect of electric field inside the active region on the electron velocity has been neglected as well as a simplified band diagram having been used for the calculations. However, even with the simplifying assumptions, a good agreement between the calculations and experiments is obtained. Obviously, these detailed processes would need to be incorporated eventually into the models of the kind under discussion here.
Figure 47. (a) Calculated ratio of the overflow electron current to the total current as a function of the EBL barrier height (\(\phi_{EBL}\)) in non-polar m-plane LEDs, assuming flat-band conditions in the active region (i.e. 0V net potential drop across the InGaN active region after the applied external voltage compensates the built-in potential, which is ~0.5V), corresponding to 3.8, 4.0, and 4.7 V externally applied bias for the LEDs with 15%, 8%, and 0% Al in the EBL, respectively. (b) Calculated ratio of overflow electron current to the total current as a function of the applied voltage (forward direction) across the m-plane LEDs with three types of EBLs: 0% Al, 8% Al and 15% Al. The symbols in (a) and (b) represent the calculated points whereas the lines are guides to the eye.

4.4d How to eliminate hot electron effects – staircase electron injector (SEI) model

Consider a DH LED but with an InGaN staircase electron injector (SEI) inserted between the n-GaN layer and the active region (In\(_{0.2}\)Ga\(_{0.8}\)N). The SEI consists of several In\(_y\)Ga\(_{1-y}\)N layers: their In fraction (\(y\)) increases in a stepwise manner starting with a low value at the first step near the junction with n-GaN. Since the conduction band discontinuity is small at the first step, the additional kinetic energy and velocity acquired by an injected electron are reduced. As a result, the probability to avoid scattering decreases while the probability to be scattered increases. Therefore, the electrons would be thermalized more efficiently by interacting with LO-phonons, and the electron overflow would be decreased or very likely eliminated. In order to quantify the
extent of the reduction in the amount of electron overflow or its elimination by employing an InGaN-based SEI, in such a way as to provide energy steps, in one or combination of more than one, equal to or greater than the LO phonon energy, we performed first-order calculations of the electron overflow for the LEDs with and without SEI (both without EBL) under different forward voltages across the active region. For simplicity of calculation and also a clearer demonstration, a one-layer SEI (with one intermediate layer of In\textsubscript{0.10}Ga\textsubscript{0.90}N) was employed (see the inset of Figure 48). The entire SEI region simulated has a total thickness of 15nm, which is the same as the total thickness of the three-layer staircase region of the LEDs investigated experimentally. An SEI with higher number of steps, as was done in our experiments, will not change the nature of this discussion.

The probability of a ballistic electron transport in the LED with one-layer SEI is given by:

\[
P_3 = \frac{\int_0^{\infty} f(E)N(E)\exp\left(-\left[\frac{d}{v_1(E + \Delta E_{c1})} + \frac{L}{v_2(E + \Delta E_{c2})}\right]\right) dE}{\int_0^{\infty} f(E)N(E) dE}
\]

Equation 16

where \(d=15\text{nm}\) and \(L=6\text{nm}\) are the thicknesses of the staircase region (In\textsubscript{0.10}Ga\textsubscript{0.90}N) and the active region, respectively, and \(v_1\) and \(v_2\) are the electron velocities in the staircase region (In\textsubscript{0.10}Ga\textsubscript{0.90}N) and the active region, respectively. The energies, \(\Delta E_{c1}\) (\(-0.25\text{ eV}\) in this case) and \(\Delta E_{c2}\) (\(-0.5\text{ eV}\) in this case) represent the excess energy gained by electrons from the conduction band discontinuities upon injection into the In\textsubscript{0.10}Ga\textsubscript{0.90}N staircase layer and cascading down to the In\textsubscript{0.20}Ga\textsubscript{0.80}N active layer, respectively. For simplicity, we assume that the electrons move in the normal direction to the hetero-interfaces. However, this assumption does not have a significant effect on the estimated probability. In particular, the electron thermalization would be
enhanced when some electrons moved at an angle with the normal direction. As a result, the actual SEI acts better than its model, and the assumed model is a good one to begin with.

In calculating the percentile of the overflow current due to the electrons that experience one scattering event we considered four different cases: *only one phonon emission or absorption in each of the staircase and the active regions*. As an example, the total probability of an electron overflow via “*only one phonon emission in the staircase region*” is composed of the following four components:

(a) Probability to find an electron suitable for this process (with energy $\hbar \omega_{ph}$ higher than the bottom of conduction band of n-GaN, as an example, for the case of no EBL and flat-band case), which is described by $P_a$ in Equation 9.

(b) Probability to reach $x$ without being scattered, which is described by $P_b$ in Equation 10, except $\Delta E_{c1}$ in place of $\Delta E_c$.

(c) Probability to emit a phonon between $x$ and $x+dx$, which is described by $P_c$ in Equation 11, except $\Delta E_{c1}$ in place of $\Delta E_c$.

(d) Probability to reach $x=d_1$ without being scattered

$$P_d = \exp \left( -\left[ \frac{(d - x)}{\nu_1 (E + \Delta E_{c1} - \hbar \omega_{LO})} \right] / \tau_w \right)$$

**Equation 17**

where $\nu_1 (E + \Delta E_{c1} - \hbar \omega_{LO}) = \sqrt{2(E + \Delta E_{c1} - \hbar \omega_{LO}) / m_e}$.

(e) Probability to reach $x = d+L$ from $x = d$ without being scattered:

$$P_e = \exp \left( -\left[ \frac{L}{\nu_2 (E + \Delta E_{c2} - \hbar \omega_{LO})} \right] / \tau_w \right)$$

**Equation 18**

where $\nu_2 (E + \Delta E_{c2} - \hbar \omega_{LO}) = \sqrt{2(E + \Delta E_{c2} - \hbar \omega_{LO}) / m_e}$.

Therefore the probability of only one phonon emission in the staircase region is given by:
\[
P_n = \left( \int_0^{E_{\text{max}}} f(E) N(E)\,dE \right) \left( \int_{\max [0, (E + \hbar \omega_{LO})]}^{E_{\text{max}}} f(E) N(E)\,dE \right) \frac{\exp \left( -\frac{[x / v_1 (E + \Delta E_{c1})]}{\tau_{\text{en}}} \right)}{v_1 (E + \Delta E_{c1}) \cdot \tau_{\text{en}}} \times \exp \left( -\frac{(d - x) / v_1 (E + \Delta E_{c1} - \hbar \omega_{LO}) + L / v_2 (E + \Delta E_{c2} - \hbar \omega_{LO})}{\tau_{\text{en}}} \right) \cdot dx \cdot dE
\]

**Equation 19**

The probability of only one phonon emission in the active region is given by:

\[
P_n = \left( \int_0^{E_{\text{max}}} f(E) N(E)\,dE \right) \left( \int_{\max [0, (E + \hbar \omega_{LO})]}^{E_{\text{max}}} f(E) N(E)\,dE \right) \frac{\exp \left( -\frac{[d / v_1 (E + \Delta E_{c1})]}{\tau_{\text{en}}} \right)}{v_2 (E + \Delta E_{c2}) \cdot \tau_{\text{en}}} \times \exp \left( -\frac{(x - d) / v_2 (E + \Delta E_{c2}) + (d + L - x) / v_2 (E + \Delta E_{c2} - \hbar \omega_{LO})}{\tau_{\text{en}}} \right) \cdot dx \cdot dE
\]

**Equation 20**

Similarly, one can obtain the probabilities for one phonon absorption events as well. The extent of the total electron overflow is obtained by summing the electron overflow due to both ballistic electrons (no scattering) and quasi-ballistic electrons (one scattering event). The contribution to overflow by electrons experiencing two or more scattering events can safely be neglected, as the step in SEI considered here is 0.25 eV below the bottom of the p-GaN conduction band and the electrons thermalized in the SEI (after multiple scattering events) have a very low probability to escape the active region. These multiple scattering events however should be taken into consideration for a more accurate calculation especially for the case of a shallow-step SEI at high injection levels.

Figure 48 shows the calculated percentile overflow electron current, namely the overflow electron current divided by the total current for two \(m\)-plane DH LEDs without EBL: one with and one without the SEI structure. The overflow current is 56%-80% of the total current within the range of applied voltages of 4V to 16V for the LED without any SEI, which is ~3-5 times larger than that of the LED with a one-layer InGaN SEI. It should be noted that for the flatband case, the electron overflow percentile is reduced to 8% with the inclusion of the particular SEI, which is still smaller than that (18% - not shown) for an LED with an increased active layer
thickness, 6 nm + 15 nm, but without any SEI (i.e. SEI step chosen to be 0.5 eV below the bottom of the p-GaN conduction band). These results suggest that for the one-intermediate layer SEI case, a sufficiently large step height and a larger SEI thickness will reduce the electron overflow substantially if not totally eliminate it.

It should be pointed out that more layers in the SEI (with potential steps no less than one LO phonon energy), like in our experiments (3-layer SEI), might provide additional freedom in the design of such structures and result in smaller electron overflow percentile. However, growth related issues in terms of the effect of SEI on the material quality and strain inside the active region\cite{106,107} should also be taken into consideration when optimizing the SEI layer stack for maximizing the LED efficiency.

![Graph showing overflow current / total current as a function of applied voltage on the LED. Symbols represent calculated values and solid lines are guides to the eye. The inset shows schematics for the LED with and without (dashed line) the SEI. For simplicity, a one-layer SEI (one In\textsubscript{0.10}G\textsubscript{0.90}N intermediate layer) was employed for the calculations.](image)

**Figure 48.** Calculated (overflow electron current / total electron current) as a function of the applied forward voltage on the LED without staircase and without EBL, and the LED with two-step InGaN staircase and without EBL. Symbols represent the calculated values and solid lines are guides to the eye. The inset shows the schematics for the LED with and without (dashed line) the SEI. For simplicity, a one-layer SEI (one In\textsubscript{0.10}G\textsubscript{0.90}N intermediate layer) was employed for the calculations.
4.4e Optimum design of SEI for minimum electron overflow

To recap, in order to smoothly segue into the design optimizations, incorporation of SEI causes the electrons to thermalize before entering the active region and in the process radically reduces if not totally eliminate the electron overflow. The question we would like to focus here is the design of the SEI structure for optimum impact. For the one-layer SEI, meaning one intermediate composition before entering the active region, smaller step height (conduction band discontinuity between the SEI and n-GaN, $\Delta E_c$ as shown in the schematic in Figure 49) could results in lower electron velocity in the SEI region therefore larger probability of being thermalized. However, a smaller step height between the n-side and the subsequent InGaN layer would result in larger conduction band discontinuity between that intermediate InGaN layer and the active region. Consequently, the probability of electrons traversing the active region without recombining would increase. As stated, in this section we will optimize the step height ($\Delta E_c$) and thickness ($d$) (see Figure 49) for the one intermediate-layer SEI with the goal of minimizing the electron overflow when no EBL is employed. Note that in section 4.4d (regarding the theoretical model of the one-layer SEI), the overflow contributed by the electrons experiencing two or more scattering events was not taken into account for a simpler treatment. However, those multiple scattering events should be taken into consideration for a more accurate calculation especially for the case of a shallow-step SEI at high applied voltages [see process “c)” in the schematic in Figure 49]. In other words, the contribution of the overflow current due to the ballistic or quasi-ballistic transport across the active region from the thermalized electrons within SEI is significant even with the reduced barrier height at the p-side by the applied voltage. It should be noted that in the theoretical model described in section 4.4d, the step in SEI is 0.25eV below the
bottom of p-GaN conduction band which is sufficiently large for the contribution of the electron overflow due to the thermalized electrons from the SEI to be safely neglected.

The electrons which obey neither the ballistic (no scattering) nor the quasi-ballistic transport \( (i.e. \) only one phonon emission or one phonon absorption) within the SEI region will be considered as thermalized down to the bottom of the conduction band of the SEI layer. The percentile of these thermalized electrons [see process “b)” in the schematic in Figure 49] can be described as \( (1-P_{bal,1} - P_{abs,1} - P_{em,1}) \), where \( P_{bal,1} \), \( P_{abs,1} \) and \( P_{em,1} \) are the probabilities of ballistic transport \( (i.e. \) undergoing no scattering), and quasi-ballistic transport with one phonon absorption and one phonon emission within the SEI region, respectively. After being injected from the SEI into the active region, these thermalized electrons will have a certain probability to transport ballistically or quasi-ballistically across the active region, depending on the conduction band discontinuity between the SEI and the active region and also the voltage drop across the active region (see process “c)” in the schematic in Figure 49). This could be described as \( (P_{bal,2} + P_{abs,2} + P_{em,2}) \), where \( P_{bal,2} \), \( P_{abs,2} \) and \( P_{em,2} \) are the probabilities of ballistic transport and quasi-ballistic transport with one phonon absorption and one phonon emission within the active region, respectively. Therefore, the amount of overflow due to the thermalized electrons from the SEI can then be described as \( (1-P_{bal,1} - P_{abs,1} - P_{em,1}) * (P_{bal,2} + P_{abs,2} + P_{em,2}) \) which is added to the overflow percentile calculated with the SEI theoretical model as described in section 4.4d to obtain the total electron overflow percentile. The calculation of these probabilities is similar to those described in section 4.4c.
Figure 49. A schematic for the conduction band of a LED with a one-intermediate layer SEI (of thickness $d$ and step height $\Delta E_c$). After being injected into the SEI from the n-GaN region, some electrons will have ballistic and quasi-ballistic (only one phonon emission or absorption) transport through the SEI [process a)], while the others (experiencing two or more scattering events) are considered to be thermalized in the SEI [process b)]. Under high bias condition and with a small step height $\Delta E_c$, the thermalized electrons from process b) might contribute to the overflow current through ballistic or quasi-ballistic transport [process c)]. For the calculations, the conduction band discontinuity between the active region and p-GaN is assumed to be 0.5eV, and no EBL is employed.

Table 2. Calculated electron overflow percentiles for a one-layer SEI, with varying SEI step height ($\Delta E_c$) and SEI thickness ($d$).

<table>
<thead>
<tr>
<th>$\Delta E_c$ (eV)</th>
<th>$d = 3 \text{ nm}$</th>
<th>$d = 9 \text{ nm}$</th>
<th>$d = 15 \text{ nm}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>36% (70%)</td>
<td>18% (66%)</td>
<td>11% (61%)</td>
</tr>
<tr>
<td>0.2</td>
<td>37% (52%)</td>
<td>18% (38%)</td>
<td>11% (21%)</td>
</tr>
<tr>
<td>0.3</td>
<td>39% (51%)</td>
<td>21% (36%)</td>
<td>11% (22%)</td>
</tr>
<tr>
<td>0.4</td>
<td>41% (52%)</td>
<td>24% (39%)</td>
<td>13% (26%)</td>
</tr>
<tr>
<td>0.5</td>
<td>42% (53%)</td>
<td>26% (42%)</td>
<td>18% (29%)</td>
</tr>
</tbody>
</table>

The calculated percentiles of electron overflow for the one-intermediate layer SEI with varying step height ($\Delta E_c$) and thickness ($d$) are shown in Table 2. Under the flat-band condition (i.e. the net potential drop across the active region $V=0$), the 15 nm-thick SEIs with step heights
of 0.1, 0.2, and 0.3eV all result in a minimum overflow percentile of 11% among the cases included in the table. It should be noticed that the 0.5 eV step height case, which basically corresponds to an increased active region thickness that would enhance thermalization of electrons within the active region, results in an increased electron overflow percentile of 18%. Under the bias corresponding to V=0.1V drop across the active region, the overflow for the 15 nm-thick SEI having a step height of 0.1eV increases to 61% due to a significant increase of overflow contribution from the electrons thermalized in the SEI but traversing the active region ballistically and or quasi-ballistically without recombination since the applied bias lowers the conduction band of p-GaN to the same level as that of the SEI. For the same bias condition corresponding to V=0.1V, the SEI structures with the step heights of 0.2eV or 0.3eV and a thickness of 15nm have comparable electron overflow percentiles (21% vs. 22%), and therefore, both could be regarded as the optimum one-layer SEI design for this particular SEI thickness. However, at higher applied voltages (e.g. V=0.2V) across the active region, it is expected that a 0.3eV or larger step height will yield a minimum overflow percentile. These results suggest that for the one-intermediate layer SEI case, a sufficiently large step height and a larger SEI thickness will reduce the electron overflow (an optimum step height for the SEI would provide a balance between the gained electron kinetic energy in the staircase region and the overflow contribution from the electrons thermalized within the SEI). However, growth related issues in terms of the effect of SEI on the material quality and strain inside the active region\textsuperscript{108,109} should also be taken into consideration when optimizing the SEI layer stack.

We now turn our attention to the case of multiple-layer SEI. Let us assume a constant SEI thickness and also a constant conduction band discontinuity between the active region and the SEI. Note that more steps within the SEI would result in smaller step height and thus less gained
kinetic energy for the electrons from the potential and thus reduced electron overflow, providing that the energy steps are equal to or larger than the LO phonon energy. As an example, we will compare the electron overflow percentile from a one-layer SEI structure and a two-layer SEI structure under the flat-band condition (see Figure 50). For both SEIs, the total SEI thickness is kept at 9nm, and the conduction band discontinuity between the active region and the SEI is 0.2eV (i.e. the step height for the one-layer SEI is $\Delta E_c = 0.3eV$). The overflow percentile for the one-layer SEI case is 21% (according to Table 2) and 16 % for the two-layer SEI. The value for the two-layer SEI was calculated taking into account the overflow electron contribution from the electrons thermalized in the SEI. The overflow percentile due to the ballistic and quasi-ballistic electron transport from point A to point C (refer to Figure 50) is 16%. A total of 19% of the electrons are thermalized and captured by the first SEI layer and the probability of ballistic and quasi-ballistic transport of these thermalized electrons from point B to point C (refer to Figure 50) is 0.8%, The product of the two probabilities (19% * 0.8%) representing the overflow gives a negligible electron overflow percentile of ~0.2%. The electron overflow contributed by the thermalized electrons in the second SEI layer is expected to be even smaller and therefore can be safely neglected. The total overflow percentile for the two-layer SEI is (16%+0.2%)≈16%, which is mainly due to the ballistic and quasi-ballistic electron transport from point A to point C. Therefore, it is reasonable to suggest that more layers in SEI (with potential steps no less than one LO phonon energy) would result in smaller electron overflow percentile. Again, the SEI effect on the material quality and strain of the active region should also be taken into consideration while maximizing the LED efficiency.
Figure 50. Schematic for a one-layer SEI and a two-layer SEI. For the one-layer SEI, the total SEI thickness is 9nm and the step height is 0.3eV. For the two-layer SEI, each step height is 0.15eV and each layer thickness is 4.5nm.

4.4f Effect of polarization field on the efficiency droop

Since the majority of commercial InGaN LEDs are grown on the polar c-plane orientation, the polarization field must be incorporated into the above-described theoretical model to be representative of the vast majority of LEDs produced and reported. In order to calculate the electron overflow in c-plane LEDs, it is necessary to consider the polarization field-induced band bending in the InGaN active region and EBL to obtain the potential energy differences of the conduction bands of n-GaN (where the electrons are injected) and EBL under various bias conditions. Again, we accomplished this task with the aid of Silvaco-Atlas commercial software package. For the simulation, polarization charge densities of $4 \times 10^{12} \text{cm}^{-2}$ and $1 \times 10^{13} \text{cm}^{-2}$ have been used for the InGaN active region and EBL with 15% Al, respectively, which is 60% of the theoretical prediction in order to facilitate the numerical convergence of Silvaco-Atlas. The other parameters are the same as those enumerated in section 4.4b. Figure 51 shows the simulated conduction band edge profile for a c-plane DH LED with 6nm active region and 15% Al in EBL, along with the m-plane counterpart under the same injection current for comparison. Due to the polarization induced field in the AlGaN EBL as well as the InGaN active
region in the $c$-plane case, the conduction bandedge of EBL is “pulled down” at the active-region side (due to the immobile positive polarization charge at the active region/EBL interface) even at the 1200Acm$^{-2}$ current density. It is for the time being assumed that the thermionic emission is the mechanism for the electrons (those that have traversed the active region by ballistic or quasi-ballistic transport) to surpass the EBL, which has a probability proportional to $\exp(-V_{EBL}/kT)$, where $V_{EBL}$ is the potential drop in the EBL (see Figure 51). The total electron overflow percentile is obtained by the product of the probability for electrons traversing the active region and the thermionic emission term $\exp(-V_{EBL}/kT)$. It should also be noted that when compared to its $m$-plane counterpart (dotted line), the polarization field in the $c$-plane LED (solid line) reduces the effective barrier of the EBL for the electrons that have traversed the active region, which suggests a higher probability for electrons to escape the active region in the $c$-plane LEDs. Therefore, an approach such as the incorporation of SEI would be needed even more so to cause efficient electron thermalization.

![Figure 51](image_url)

**Figure 51.** Calculated conduction band structures of $c$-plane (solid line) and $m$-plane (dotted line) InGaN LEDs with a 6nm In$_{0.15}$Ga$_{0.85}$N active region and a 10nm Al$_{0.15}$Ga$_{0.85}$N EBL. The injection current density for both LEDs is 1200Acm$^{-2}$. 

111
Figure 52. Calculated electron overflow current as a function of the applied voltage on the LED for c-plane DH LEDs (6nm In_{0.15}Ga_{0.85}N active region) with different EBL heights (0% Al and 15% Al, respectively) and without SEI as well as a c-plane DH LED with SEI and without any EBL.

The calculated percentile of electron overflow for c-plane 6nm DH LEDs is shown in Figure 52. In the case where no SEI is employed, the LED without EBL (solid circles) in overall shows a much higher electron overflow percentile than that with 15% Al in the EBL (solid squares). Moreover, the overflow in the LED without an EBL saturates at 90% once the applied voltage exceeds 5V, while for the LED with 15% Al in EBL the overflow increases with the applied voltage: from 0% to 50% when the voltage increases from 3V to 14V. For the c-plane LED which has a one-layer SEI and no EBL, the calculated electron overflow is greatly reduced (~4.5 times lower which is consistent with experiments as will soon be shown) compared to that having no SEI and no EBL. Just for reference, the simulated current densities corresponding to 5 V and 14 V externally applied bias (neglecting any voltage drop across metal/semiconductor contacts) are ~990-1000 Acm^{-2} and ~6820-6850 Acm^{-2}, respectively, for all three LEDs. Also at
5 V and 14 V applied bias, the net potential drop across the InGaN active region is, respectively, \(~0.11\text{V}\) and \(~0.05\text{V}\) for the c-plane LED with an \(\text{Al}_{0.15}\text{Ga}_{0.85}\text{N}\) EBL, \(~0.33\text{V}\) and \(~0.25\text{V}\) for the c-plane LED without any EBL, and \(~0.08\text{V}\) and \(~0.002\text{V}\) for the c-plane LED with one-layer SEI.

It is evident that the c-plane LEDs (Figure 52) have higher electron overflow than their m-plane counterparts (Figure 48) having the same structures. This is due to the polarization-induced band bending in the InGaN active and EBL regions of the c-plane LEDs which results in a reduced effective barrier provided by EBL (Figure 51). In this regard, the polarization field exacerbates the electron overflow in LEDs. Again, the responsible mechanism behind the electron overflow is the ballistic and quasi-ballistic electron transport since the EL efficiency loss caused by electron overflow is also present in the m-plane LEDs, which is free of polarization induced field. We should note that the calculations do not take into consideration the effect of acceleration of electrons under the electrical field within the active region, when band bending exists, which might have underestimated the calculated percentiles of overflow. It is worth mentioning that in the current c-plane case even without considering the acceleration of the electron by the electric field, the probability for an electron to traverse the active region and reach the EBL (before surpassing the EBL) is already \(~90\%\) (in both 0% Al and 15% Al EBL cases) due to the lowering of the barrier in the p-region by the polarization-induced electric filed. Marginal (3-5\%) enhancement of the ballistic or quasi-ballistic electron transport is gained by adding the electric field acceleration effect on the electron velocity in the present case.
**4.4g EL efficiency of InGaN LEDs with SEI**

In order to test our premise in regard to hot electrons and the ensuing ballistic and quasi-ballistic electron transit across the active region without partaking in the recombination process, (without complete thermalization and reaching equilibrium with the lattice), two *m*-plane LEDs were investigated [see Figure 53(a) for the device schematics]: one LED with a 10nm *p*-AlGaN EBL with 15% Al, while the other without any EBL at the *p*-region, and both having a SEI under the InGaN active region to cause the thermalization of the injected electrons. As will be seen below, we extended this study to include the *c*-plane variety as well.

The SEI consists of three 5nm InGaN layers with In compositions of 3%, 6% and 10%, in the order starting from the n-GaN side, as shown in Figure 53(a) (the first step height going from n-In$_{0.01}$GaN to the first SEI layer, In$_{0.03}$GaN, is very close to or smaller than an LO phonon energy (88 meV); therefore, the first two SEI layers may be considered as a single step for electron cooling via LO phonon emissions. The step heights for other steps in the SEI are more than the LO phonon energy as desired.). In order to circumvent the quality degradation by the SEI layer, a six-period In$_{0.01}$Ga$_{0.99}$N(7nm)/In$_{0.06}$Ga$_{0.94}$N(3nm) multiple quantum well (MQW) underlayer was employed prior to the active layer underneath the 12 nm-thick In$_{0.01}$Ga$_{0.99}$N layer and the InGaN SEI to improve the active region quality. The underlayer and the SEI were *n*-type doped with Si to an electron density of 2×10$^{18}$cm$^{-3}$.

Figure 53(b) shows the relative EQE of the two *m*-plane LEDs with SEI; as mentioned, one LED has the EBL and the other has not. The results confirm that the LED with the EBL has essentially the same EL intensity (i.e. relative EQE) as that without any EBL. This is substantially different from what we have observed in Figure 43(a) in regard to devices not featuring an SEI, where the EL intensity from the LED without EBL is 3-5 times lower than the
one with EBL. It is, therefore, reasonable to suggest convincingly that by employing the InGaN SEI before the active region, the electron overflow due to ballistic or quasi-ballistic electron transport across the active region is eliminated, which has resulted in a similar EL performance for the two LEDs with SEI regardless of the EBL status. This is reasonable considering that the electron overflow solely due to thermionic emission is negligible (even for the LED without EBL) according to the discussion in section 4.4b.

![Diagram of LED structure](image)

**Figure 53.** (a) Schematic for the two m-plane LEDs with InGaN SEI before the active regions (to thermalize the injected electrons from the n-GaN layers), one of which has a 10nm EBL with 15% Al and the other one without any EBL in the p-region. (b) Relative EQE of the two m-plane LEDs with a 3-layer staircase electron injector (SEI) 6nm InGaN active region λ~440nm
with staircase electron injector: one with and one without EBL, respectively. The LEDs were measured under pulsed current with a frequency of 1kHz and 1µs duty width.

Similar to the m-plane case, the significant difference between the LEDs with an EBL and without EBL is eliminated when an InGaN-based SEI structure is employed in the c-plane LED case as well. The same three-layer SEI and the MQW underlayer (for improving the active region materials quality) structures used in m-plane LEDs are employed also for the c-plane LEDs. Both c-plane LEDs with SEI under investigation, one with EBL (15% Al) and the other without any EBL, exhibit essentially the same IQE as determined from PL measurements with resonant excitation (IQE of 46%-50% at a carrier density of $1\times10^{18}$ cm$^{-3}$ assuming a radiative recombination coefficient $B$ of $1\times10^{-11}$ cm$^{3}$s$^{-1}$). Figure 54 shows that when the SEI is employed, the difference in relative EQE between the LEDs with and without EBL is significantly reduced as compared to that in LEDs without SEI in Figure 44. This once again suggests that by using the SEI the ballistic and quasi-ballistic electron transport across the active region is substantially reduced. However, the injection current dependence of the efficiency in the c-plane LEDs with SEI deviates somewhat from that of the m-plane LEDs with SEI, where essentially the same EQE values were observed for the LEDs with and without EBL for all injection current levels. Somewhat unexpectedly, for the c-plane case the LED without EBL shows even higher EQE than that with the EBL when the current density is below 1700 Acm$^{-2}$. At low current densities (<500 Acm$^{-2}$), the EQE from the LED without EBL is 1.7-2.5 times higher than the one with EBL. When the current density is above 1700 Acm$^{-2}$, the former shows slightly lower EQE values than the latter (<8%), within nominal variations due to e.g. processes such as current filamentation.

The removal of the EBL has a more significant improvement in terms of EQE for the c-plane LEDs than for the m-plane ones. This might result from a more inferior hole transport
across the EBL in the $c$-plane case due to relatively larger hole effective mass compared to that in the $m$-plane LEDs and or relatively lower hole density. Additionally, the 22% efficiency degradation observed for the LED with SEI and without EBL when the current density is increased up to 2500 Acm$^{-2}$ might suggest that the $c$-plane orientation requires more thermalization due to its polarization field in order to avoid the electron overflow-induced EL efficiency loss. Of course, other processes such as current filamentation can play their role as well.

![Figure 54. Relative EQE of two $c$-plane LEDs with SEI inserted under the active region: one with and one without EBLs, respectively. The LEDs were measured under pulsed current with a frequency of 1kHz and 1µs pulse width. The SEI includes three intermediate InGaN layers with In composition of 3%, 6% and 10%, respectively.](image)

It should be pointed out that the efficiency degradation caused by the hot electrons can either be reduced by an EBL (to block the escaping electrons) or a SEI (to thermalize the hot electrons before they are injected into the active region). However, the EBL not only acts to reduce electron overflow, but also presents a barrier for hole injection, albeit a relatively small one, which is not desirable considering that hole injection is already hampered due to relatively
low hole density on the p-side. Moreover, in the c-plane LED case (the most commonly used orientation), the polarization field inside the active region and EBL could greatly reduce the effective barrier of the EBL for the electrons.\textsuperscript{110} The employment of a SEI automatically reduces the gained kinetic energy of the injected electrons by reducing the conduction band discontinuity to about one LO phonon energy at a step. This increases the electron transit time during which the hot electrons would be more likely thermalized by LO-phonon scattering, not to mention the barrier they will face on the other side of the active region, which culminates into reducing the electron overflow.

In short, an InGaN SEI with gradually increased In composition was inserted prior to the active region to reduce the electron overflow caused by ballistic and quasi-ballistic electron transport without any EBL layer which has the deleterious effect on the hole injection and is not welcome for technological reasons. The employment of SEI resulted in the approximately same EL performance for the m-plane LEDs with and without EBL. For the polar c-plane LED case, up to a factor of 2.5 improvement has been observed in EQE for the c-plane LED with SEI but no EBL at current densities below 1700 Acm\textsuperscript{2} due possibly to enhanced hole injection since the EBL also impedes the hole transport.

4.5 Effect of current crowding on EL efficiency degradation

Another cause of efficiency loss has its genesis in the detrimental effect of current crowding. In some LEDs where a lateral contact configuration is employed, either for n- or the p-side, the current density is distributed non-uniformly across the device area. It is concentrated near the edges of the thicker metal around the mesa and decreases exponentially with distance from the mesa edge.\textsuperscript{79} While this is more detrimental in the p-layer due to its large resistance, the
n or p-layer contact configuration, depending on regular or flip-chip mount, also contributes and must be taken into consideration. In LEDs not using the flip-chip technology, meaning the p-layer is on the top and through which the light is extracted, the current is denser near the metal electrodes whose distribution and geometry must be optimized with current crowding in mind.

The current concentrated near the metal grid/contacts can result in efficiency degradation due to locally increased injection levels which results in local heating which in turn results in a higher current, a detrimental positive feedback. The particulars of current crowding in lateral contacts, although an age old problem in many devices including bipolar transistors, have been discussed in the context of nitride based LEDs in numerous papers, see, e.g., ref. 111. In addition to the contact configuration, a proper choice of p-contact material is also critical for relieving the current crowding effect, thereby reducing the extent of the efficiency degradation as supported by our comparative studies of LED structures which employed a highly conductive Ga doped ZnO (GZO) layer\textsuperscript{112} and a semi-transparent Ni/Au as p-contact materials. The GZO p-contact was grown by molecular beam epitaxy on an MQW-LED sample which was in turn grown on c-plane sapphire with six-period 2nm In\textsubscript{0.15}Ga\textsubscript{0.85}N wells and 12nm In\textsubscript{0.01}Ga\textsubscript{0.99}N barriers. The relatively thick barriers employed were to allow efficiency degradation so that a comparative analysis of the efficiency loss can be made. For comparison, semitransparent Ni/Au (5nm/5nm) p-contact was used on another piece of the same LED wafer as a reference sample. The results show that in addition to nearly doubling the light extraction due to improved transmittance of GZO and more uniform light emission over the thin Ni/Au contact, the extent of the efficiency degradation was also reduced from \(\sim 37\%\) for the LED with thin Ni/Au contact to \(\sim 27\%\) for the one with GZO contact in tests up to current levels of 3500 Acm\textsuperscript{2}.\textsuperscript{113} The reduced efficiency drop in the GZO case is attributable to minimization if not the total elimination of the current
crowding effect. Note that a much more uniform emission intensity distribution at high current levels is achieved for the LED with GZO contact layer as compared to the case with thin Ni/Au contact which exhibited non-uniform light emission at high injection levels.

4.6 Effect of junction heating on EL efficiency degradation

Last but not least, junction Joule heating effect on LED efficiency deserves a close attention as it has also been proposed to contribute to the InGaN LED efficiency drop, indicating the obvious need for optimal heat removal. The junction heating issue is very important due to several reasons. First, it affects the internal quantum efficiency due to enhanced non-radiative processes including the SRH recombination. Second, excessive junction heating would also degrade the contacts and contact/semiconductor interfaces, which would manifest itself as an increased series resistance, thereby degrading efficiency as well as the power conversion efficiency. At high currents, the dominant heat source is from Joule heating of the p-type GaN and p-contact, having a dependence of $I^2R$. Additional heat may also come from an increased non-radiative recombination due to the layer degradation at high operating currents and low photon extraction efficiency. The aforementioned discussion goes somewhat counter to that proposed by Kim et al. which stated that the junction heating only reduce the overall efficiency of the LEDs, but not the cause of the efficiency drop. According to our measured EL data, at high injection levels thermally induced drop is observed even at 1% duty cycle as judged from the fact that the drop can be partially eliminated by employing 0.1% duty cycle.
4.7 Summary

Significant progress in the power conversion efficiency and brightness of InGaN-based light emitting diodes (LEDs) has paved the way for these devices to be considered for LED lighting. In this realm, however, the efficiency must be retained at high injection levels in order to generate the lumens required. Unfortunately, LEDs undergo a monotonic efficiency degradation starting at current densities even lower than 50 A/cm² which would hinder LED insertion into the general lighting market. As described in this chapter, the loss of efficiency retention at high injection levels has so far been attributed to many phenomena which include Auger recombination, carrier overflow, large effective mass of holes which also provide an impediment to hole transport across the MQW region used, relatively low hole density in the p-layer (particularly in relation to the electron density on the n-side), defects, and current crowding effects. Using LED structures with varying EBL (0-15% Al) barriers, we determined the presence of ballistic and or quasi-ballistic electrons in the active layer and their role in efficiency reduction in general and with increasing current in particular by considering the electron scattering and transport in this new realm. Specifically, we developed first order models utilizing the LO phonon scattering rates to describe the electron overflow to the p-type region resulting in 3-5 times lower EL intensity in LEDs without the EBL compared to those with Al₀.₁₅Ga₀.₈₅N EBL both for c-plane and m-plane varieties. For an effective electron thermalization, and therefore, reduction of the ballistic and quasi-ballistic transport of the injected electrons to reduce the electron overflow responsible for the efficiency degradation in these devices, an InGaN staircase structure (with step-wise increased In composition) was inserted before the active region. The employment of the SEI resulted in essentially the same electroluminescence performance for the LEDs with and without EBL for the polarization charge void m-plane case.
For the polar c-plane LED case, up to a factor of 2.5 improvement has been observed in EQE when SEI was inserted but EBL removed for current densities below 1700 Acm\(^{-2}\) due possibly to enhanced hole injection since the EBL also impedes the hole transport. Lack of any EBL is beneficial in that any barrier presented to hole injection and transit is eliminated. Our calculation confirms that the insertion of a staircase injector structure is beneficial for a better electron thermalization and a reduced electron overflow.
Chapter 5. GaN-based vertical cavities on highly reflective and crack-free nitride distributed Bragg reflectors

5.1 Motivation

In the last couple of decades there have been tremendous advancements in nitride-based edge emitting lasers, which have now become commercially available. Lasers of the other variety, vertical-cavity surface emitting lasers (VCSELs) are very promising for high-density storage, laser printing, display applications due to their inherent advantages such as narrower beam divergence, lower threshold current, lower power consumption, higher modulation bandwidth, and easier light coupling with optical fibers when compared to their edge emitting counterparts. However, the research on nitride-based VCSELs is still in its early stages, as their development has faced a number of challenges among which is the growth of high reflectivity and crack-free nitride-based distributed Bragg reflectors (DBRs). AlN/GaN DBRs have been widely studied due to high refractive index contrast between the constituents, thereby leading to a relatively wide stopband width with a reduced number of pairs to achieve high reflectivity. However, a large built-in strain in the DBR structure and thereby a high density of cracks have been observed and are very hard to avoid due to the large lattice mismatch (2.4%). These cracks seriously affect the reflectivity of DBRs due mainly to light scattering and diffraction. Therefore, it is necessary to suppress if not totally eliminate crack generation in order to achieve high reflectivity DBRs and high finesse optical cavities. As a partial remedy to the above-mentioned cracking issues with AlN/GaN DBRs, the growth of AlGaN/(Al)GaN DBRs has been reported to mitigate the cracking issue but at the expense of increased number of pairs.
and reduced stopband width due to reduced refractive index contrast\textsuperscript{123,124,125,126,127,128,129}. The crack density could be reduced or even eliminated in some cases, but the total thickness of DBR is still limited. By using lattice-matched AlInN/(Al)GaN DBRs, the built-in strain, therefore cracking, has been eliminated\textsuperscript{130,131,132}. However, AlInN/(Al)GaN DBRs still have major drawbacks. The composition of AlInN is highly sensitive to its growth temperature, and AlInN/GaN DBR growth involves numerous temperature ramping processes that increase the total growth time (typically 12-14 hours for 40.5 pairs) since AlInN and GaN require different growth temperatures. Moreover, AlInN/GaN DBR growth also consumes relatively high amount of ammonia to obtain reasonable In incorporation efficiency in AlInN layers.

As an approach to avoid crack formation, selective growth has been employed for AlAs/GaAs DBRs, which also suffer from the cracking problem due to strain, on patterned InP heterostructures\textsuperscript{133}. This method has also been successfully applied to the epitaxial growth of GaN on Si (111) substrates to relax the tensile strain in the GaN films introduced by the large difference in thermal expansion coefficients of GaN and Si\textsuperscript{134}. In the current work, we demonstrate GaN-based vertical cavities using crack-free high reflectivity (~ 98 \%) Al\textsubscript{0.46}Ga\textsubscript{0.54}N/GaN DBRs grown selectively by metalorganic chemical vapor deposition (MOCVD) on patterned AlN templates. This technique is very promising for fabricating nitride-based VCSEL devices with crack free areas up to 150 × 150 \(\mu\)m\textsuperscript{2}, which is much larger than that used for typical VCSEL devices.

### 5.2 Experimental procedure

The AlGaN/GaN DBR samples were prepared by MOCVD, where trimethylgallium (TMGa), trimethylallium (TMAI), and ammonia were used as the Ga, Al, and N sources, respectively. After the growth of a 500 nm-thick AlN buffer layer at 1030 °C on c-plane sapphire,
the wafer was coated with a 200 nm-thick SiO₂ film using plasma-enhanced chemical vapor deposition (PECVD). Square-shaped windows with 10 μm separation were opened on the SiO₂ film to expose the underlying AlN film by using standard photolithography and wet etching of SiO₂ with buffer oxide etch (BOE). The windows of sizes 150 × 150 μm², 100 × 100 μm², 75 × 75 μm², and 50 × 50 μm² were grouped on the same wafer such that those with the same dimensions formed arrays having areas of 2.2 × 2.2 mm². The patterned window edges were aligned along the GaN<11̅20> or <11̅00> directions. A 40.5 pair patterned Al₀.₄₆Ga₀.₅₄N/GaN DBR was then grown at 990 °C with 2 SLM (standard liter per minute) NH₃ flow rate at a growth pressure of 30 Torr. The total growth time was approximately 6 hours. The as-grown DBR samples were characterized by scanning electron microscopy (SEM), high-resolution x-ray diffraction (XRD), and optical reflectivity measurements. A 6λ cavity layer which consists of five periods of In₀.₀₁Ga₀.₉₉N/In₀.₂₀Ga₀.₈₀N QWs was deposited on the patterned Al₀.₄₆Ga₀.₅₄N/GaN DBR, with its central quantum well aligned at a position that is 4λ (λ=440nm) away from the cavity/bottom DBR interface and 2λ away from the cavity/top DBR interface. The full cavity structure was completed with the deposition of a 13 pair SiO₂/SiNx DBR on top by PECVD, which had a peak reflectivity of ~99.5% and a stop-band width of ~90 nm.

5.3 Results and discussion

For a 40.5 pair patterned AlGaN/GaN DBR, the surface was crack-free across a whole 2 inch wafer, as shown in Figure 55(a). There is some deposition with random orientation in the masked area as shown in Figure 55(b) due to the poor growth selectivity of AlGaN. The gap width between each square window shows negligible change after the DBR growth, suggesting negligible lateral overgrowth of the DBR under the particular growth conditions employed. The cross-sectional SEM image of Figure 55(c) shows a clear contrast between the individual GaN
(dark) and AlGaN (bright) layers. As in the case of planar DBRs, the AlN buffer layer plays a very important role in releasing the tensile strain built in the patterned AlGaN/GaN DBRs.\textsuperscript{135}

High-resolution XRD measurements were carried out on the patterned DBR sample to confirm the Al composition and the pair thickness. The x-ray spot size on the sample was reduced to 1 mm × 1 mm, which is smaller than the size of the area containing the square patterns of the same dimension (2.2 × 2.2 mm\textsuperscript{2}). Figure 56 shows the XRD θ-2θ scan from an area containing 150 × 150 μm\textsuperscript{2} patterns. The diffraction peaks from GaN, AlGaN, and AlN are clearly resolved with thickness oscillations superimposed. Al composition in AlGaN layer was estimated as ~46 %. The total thickness of a single pair of Al\textsubscript{0.46}Ga\textsubscript{0.54}N/GaN was calculated as 94 nm from the separation of these oscillation peaks in good agreement with the designed thickness (GaN: 44.7 nm, Al\textsubscript{0.46}Ga\textsubscript{0.54}N: 49.5 nm). Similarly, the pair thickness for an area with 50 × 50 μm\textsuperscript{2} patterns was estimated as 102 nm. This higher growth rate of DBR with reduced pattern size is due to availability of relatively more source material resulting from decreased proportion of the total growth area to the total masked area.
Figure 55. (a) A plan-view SEM image of a selectively-grown 40.5 pair $\text{Al}_{0.46}\text{Ga}_{0.54}\text{N}/\text{GaN}$ DBR, with a pattern size of $150 \times 150 \mu\text{m}^2$. Each square is separated from each other by a 10 $\mu\text{m}$ wide $\text{SiO}_2$ mask. DBR Surface is crack-free across the whole 2 inch wafer. (b) A close-up plan-view SEM image of the DBR sample near the $\text{SiO}_2$ mask at the pattern edge. (c) A cross-sectional SEM image of the selectively grown DBR sample showing individual GaN (dark) and AlGaN (bright) layers in the DBR.

Figure 56. XRD $\omega$-20 scan of the AlGaN/GaN DBR with pattern size of $150 \times 150 \mu\text{m}^2$. From the clearly observed fringes the DBR pair thickness was determined as $\sim 94$ nm.
Optical reflectivity measurements at room temperature were conducted on the AlGaN/GaN DBR sample at the normal incidence using a Xenon lamp and a micro-objective lens (50×) to probe a single DBR window. The data were calibrated by using the reflectivity spectra of a UV-enhanced Al mirror. As shown in Figure 57 the peak reflectivity values range from 96.0 % to 98.0 % for DBR windows with different dimensions located near the center of a 2 inch wafer. The stopband width for all pattern sizes is ~ 14 nm, which is smaller than the stopband width of ~22 nm calculated using the scattering matrix method\textsuperscript{136}. This discrepancy is most likely due to thickness and composition (\textit{i.e.} refractive index) fluctuations in the AlGaN/GaN DBR. The stopband center wavelength varies from 440 nm for the largest pattern size (150 × 150 μm\textsuperscript{2}) to 470 nm for the smallest pattern size (50 × 50 μm\textsuperscript{2}), due to the increasing growth rate with decreasing pattern size as described above. It should be noted that measurements were performed on neighboring DBRs of different pattern sizes to avoid the effects of nonuniformity across the wafer.
Figure 57. Reflectivity spectra from the DBR samples with different pattern sizes. With the decrease of pattern size, the stop bands blue shift, suggesting higher growth rate of DBR due to the selective growth.

Figure 58. Schematic for a full micro-cavity structure on patterned a 40.5 pair patterned Al$_{0.46}$Ga$_{0.54}$N/GaN DBR.

The full vertical cavity structure (shown in Figure 58) consists of cavity layer having 5 InGaN/InGaN MQWs sandwiched between the bottom AlGaN/GaN DBR and the top SiO$_2$/SiN$_x$ DBR, the middle quantum well is located $2\lambda$ ($\lambda=440 \text{nm}$) away from the top surface of the cavity, and $4\lambda$ away from the bottom of the cavity so that the middle quantum well is at the anti-node position and the optical gain will be maximized. Micro-photoluminescence (PL) measurements at room temperature were conducted to investigate the cavity modes. A frequency-doubled 80 MHz mode-locked femtosecond Ti:Sapphire laser emitting at 380 nm was used as the excitation source in the transmission configuration. The beam was focused into a circular spot with a diameter of approximately 5 $\mu$m using a micro-objective lens. Figure 59 shows the typical micro-PL spectrum at room temperature from a $150 \times 150 \mu$m$^2$ pattern size device. The full-cavity
exhibits a PL emission peak at 442 nm and a quality value (Q-value) of ~ 300. It should be noted that there is a position dependence across the wafer for both the bottom DBR reflectivity and the cavity layer thickness due to spatial nonuniformity in the growth rate. Further optimization is needed to increase the top and bottom DBR reflectivities, achieve sharper cavity interfaces and better thickness control, and reduce absorption loss, all of which are required to obtain higher Q-value that would lead to lasing action.

![Figure 59](image)

**Figure 59.** Room-temperature micro-photoluminescence spectra (solid line) from full-cavity structure measured from the back surface of the sample. The dashed line represents the reflectivity data from the bottom DBR, which is a 40.5 pair patterned Al_{0.46}Ga_{0.54}N/GaN DBR. The top DBR is a 13 pair SiO_{2}/SiN_{x} DBR, which has a peak reflectivity of ~99.5 %. The sample was excited by a frequency-doubled Ti:Sapphire laser emitting at 380 nm from the top DBR side. The device pattern size is 150 × 150 μm². The Q-value deduced from the PL spectrum (λ/Δλ) is approximately 300.

### 5.4 Conclusion

A crack-free Al_{0.46}Ga_{0.54}N/GaN DBR with ~98% reflectivity was selectively grown with square patterns of up to 150 × 150 mm² in size with MOCVD. Vertical cavity structures were obtained on these crack-free patterned DBRs. A cavity mode at ~ 442 nm in 150 × 150 mm² area
was observed, having quality factor \(\sim 300\). Even though further optimization is needed to achieve lasing action, the selective growth technique was shown to be very promising for high quality nitride DBRs, and therefore, for GaN-based VCSEL devices.
Chapter 6. Future work

6.1 Preparation of large-area high-quality m-plane substrates

First, let’s go back to the GaN template preparation again. As we have demonstrated in Chapter 3, m-plane GaN is promising for optoelectronic applications due to the lack of polarization-induced electric field and additionally due to a reduced valence band effective mass (thus smaller acceptor binding energy), and larger optical matrix elements relative to its c-plane counterpart. In this thesis, we investigated the preparation of high-quality m-plane non-polar GaN with epitaxial lateral overgrowth (ELO) method on both sapphire and Si substrates. Especially the demonstration of m-plane GaN growth on patterned Si substrate is of a great promise in that it is a method to obtain low-cost large-area high-quality m-plane GaN substrate for optoelectronic device applications. However, further improvement is needed for this method. It has been found that a limited number of cracks separated by hundreds of microns along the c-axis caused by the large difference of thermal expansion coefficient between GaN and Si. This cracking will occur during wafer cooling stage, and will cause serious meltback reaction between Ga and Si when the wafer is loaded back into the MOCVD system for regrowth, which consequently results in very rough surface of GaN layer and also the formation of etched pits inside the Si substrates. In order to solve this cracking and meltback issue, the buffer layer between the GaN epilayer and Si substrates need to be optimized. Possible solutions include using low-temperature AlN\textsuperscript{57,58} or AlN/GaN superlattices\textsuperscript{59} as a buffer layer to release the thermally-generated strain inside the GaN film, thereby to avoid the formation of cracks. For the case of a low-temperature AlN buffer, typical growth temperature of 700-800°C with a thickness of 30nm have been reported.\textsuperscript{57,58} For the AlN/GaN superlattices as a buffer layer, the optimum
thickness for the superlattices is 3nm and 4 nm for the AlN and the GaN, respectively at a growth temperature of 1060 °C.\textsuperscript{59}

### 6.2 Hot-electron overflow model

As we have discussed in chapter 4, a very simplified first-order model has been used to demonstrate the electron overflow phenomena caused by the hot electron transport (or ballistic and quasi-ballistic transport). However, the hot electron overflow model has made certain simplifications even though these simplifications do not change the nature of the discussion: (1) not including the electric field effect on carrier velocity, (2) using simplified band diagram, (3) not including the dependence of phonon lifetime on the carrier density\textsuperscript{104}, etc. Unfortunately, the carrier distribution inside the active region is affected by the carrier overflow caused by the hot electrons, while the carrier overflow is also dependent on the carrier distribution inside the active region. Therefore, a more accurate and comprehensive calculation would require that the incorporation of the hot electron overflow equations into the device simulation software, and should be solved together with the Poisson’s equations, carrier continuity equations, and drift-diffusion equations. However, at the moment major commercial simulation packages such as Silvaco Atlas®, APSYS®, SiLENSe®, are lacking this hot electron overflow mechanism, which would certainly compromise their ability in accurately predicting LED performance especially at high injection currents where electron overflow is significant. A collateral effort is needed from both our lab and simulation package developer in order to realize the incorporation of hot electron overflow model into the device simulation packages.
6.3 Enhancement of hole injection into active region

By the employment of staircase electron injector (SEI), the hot electron overflow could be significantly reduced if not totally eliminated. However, one should keep in mind that the electrons need to be recombined with holes to generate photons, meaning that the enhancement of hole injection into the active region could be important to the LED EL performance as well considering that holes concentration is typically lower than electron concentration. A technologically promising way to enhance the p-type doping is Mg $\delta$-doping, as reported in Refs 137 and 138. It is demonstrated that Mg $\delta$-doping (as shown in the schematic in Figure 60) enhances the vertical (five-fold increase) and lateral (two-fold increase) conductivities, as well as the materials quality of p-GaN due to a decrease in Mg activation energy owing to dense packing of Mg dopants.138 On the other hand, Mg-doped AlGaN/GaN superlattice structure has also been reported by several groups to enhance the hole conduction in p-layer.139,140 However, the enhancement of hole injection by employing such a superlattice structure is very limited because the structure also introduces potential barriers for hole injection in the vertical direction. For Mg $\delta$-doping, the major parameters for the growth process include the p-layer growth temperature (affecting the Mg diffusion into u-GaN layers), Mg flux (determined by the Cp$_2$Mg flow rate $f_{Mg}$ and deposition time $t_{Mg}$), and so on.
Figure 60. A schematic for Mg δ-doping. The doping parameters include the nominally undoped GaN layer thickness $d$, and Mg flux $P_{Mg}$ (determined by the Cp$_2$Mg flow rate $f_{Mg}$ and deposition time $t_{Mg}$) with NH$_3$ flow at the same time. (after Ref. 138)
Appendix A: IQE from power dependent photoluminescence measurement

It is assumed that at steady state the total generation rate \( G \) is equal to the total recombination rate \( R \) which includes Shockley-Read-Hall nonradiative recombination \( (A_n) \), bimolecular radiative recombination \( (B_n^2) \), and Auger recombination \( (C_n^3) \) if any, where \( n \) is the carrier concentration, \( i.e. \)

\[
G = A_n + B_n^2 + C_n^3
\]

Equation 21

The measured PL intensity could be represented as

\[
I_{PL} = \eta_c B n^2
\]

Equation 22

where \( I_{PL} \) is the integrated PL intensity, the collection factor \( \eta_c \) includes escape efficiency of photons as well as the collection efficiency of luminescence by the optics/detector, which is constant during a given measurement but different from measurement to measurement even though attempts are made to keep the collection geometry the same.

By eliminating \( n \) from Equation 21 & Equation 22, one could obtain

\[
G = \frac{A}{\sqrt{\eta_c B}} \sqrt{I_{PL}} + \frac{1}{\eta_c} I_{PL} + \frac{C}{(\eta_c B)^{3/2}} \left( \sqrt{I_{PL}} \right)^{3/2}
\]

Equation 23

Here \( G \) is a function of \( \sqrt{I_{PL}} \). By fitting the plot of \( G \) vs. \( \sqrt{I_{PL}} \), one could obtain the three fitting coefficients: \( P_1 = \frac{A}{\sqrt{\eta_c B}}, P_2 = \frac{1}{\eta_c}, P_2 = \frac{C}{(\eta_c B)^{3/2}} \). Fortunately \( G \) could also be calculated separately from experimental parameters:
\[ G = \frac{P_{\text{laser}}}{R} \frac{(1-R)\alpha}{A_{\text{spot}} h\nu} \]

**Equation 24**

where \( P_{\text{laser}} \) is the optical power incident on the sample, \( R \) is the Fresenel reflection at the sample surface, \( A_{\text{spot}} \) is the laser spot size, \( h\nu \) is the energy of a photon from laser source, \( \alpha \) is the absorption coefficient of the InGaN active layers at the laser wavelength.

When the generated stead-state carrier density \( n \) is relatively low, the Auger recombination term is very weak as compared to the radiative recombination term (i.e. the \( An^3 \) term is much smaller than \( Bn^2 \) when \( n \) is usually in \( 10^{17}-10^{18}\text{cm}^{-3} \)), and then Equation 23 could be simplified to:

\[ G = \frac{A}{\sqrt{\eta_c B} \sqrt{I_{\text{pl}}} + \frac{1}{\eta_c I_{\text{pl}}}} \]

**Equation 25**

The absorption coefficient \( \alpha \) of InGaN can be obtained by

\[ \alpha = \alpha_0 \sqrt{\frac{(E-E_g)}{E_g}} \]

**Equation 26**

where \( \alpha_0 \) is the absorption coefficient at \( h\nu = 2E_g \), is obtained by linear interpolation between GaN and InN (\( \alpha_0 = 2.0 \times 10^5\text{cm}^{-1} \) for GaN, \( 1.2 \times 10^5\text{cm}^{-1} \) for InN).

At each laser excitation power, one could obtain \( G \) and \( I_{\text{pl}} \). As mentioned above, by fitting the plot of \( G \) vs. \( I_{\text{pl}} \), one could obtain the coefficients \( P_1, P_2 \) & \( P_3 \).

Then the IQE could be calculated by

\[ \text{IQE} = \frac{Bn^2}{G} = \frac{I_{\text{pl}}}{\eta_c G} = \frac{I_{\text{pl}} P_2}{G} \]

**Equation 27**

Knowing \( I_{\text{pl}}, G \) and \( P_2 \), the IQE value at each laser excitation power (or generation rate) could be obtained.
It should be noted that a constant $B$ parameter has been assumed with the increase of laser excitation power. However, this assumption is not true due to two reasons: i) in $c$-plane LEDs the screening of polarization-induced electric field with the increase of injected carriers could change the radiative recombination coefficient $B$; ii) the formation and dissociation of excitons cannot be neglected even at room temperature given an exciton binding energy of the order 20meV, also leading to a strong change in $B$ with injected carrier density inside the active region.

As an example, Figure 61 shows the fitting of the generation rate $G$ as a function of the integrated PL intensity $I_{PL}$ for a $c$-plane LED active layer (with 6 nm InGaN DH) on sapphire (the reference sample used in figure 31 in section 2.4b). During the calculation of $G$ values, a diameter of 200µm has been used to estimate the laser spot size. Measurement was carried out on the LED active layer sample at room temperature using a frequency-doubled 80 MHz repetition rate femtosecond Ti:Sapphire laser. The excitation laser wavelength was 370 nm, below the bandgap of the quantum barriers and top GaN.

With the obtained $P_z$ and Equation 27, we could obtain IQE value with as a function of generation rate (i.e. laser excitation powers) without knowing the values of $A$ and $B$ coefficients. After assuming (or measuring) a value for $B$ coefficient ($1 \times 10^{-11}$ cm$^3$s$^{-1}$ used here), one could obtain the steady-state carrier density $n$ by $IQE = \frac{Bn^2}{G}$, thereby the IQE values as a function of the generated carrier density $n$, as shown in Figure 62. Alternatively, the IQE values as a function of $n$ could also be obtained using the procedures described in ref. 16: by solving the equation
\[ G = P_1 \sqrt{\eta_c} \sqrt{Bn} + \left( \sqrt{Bn} \right)^2 \]

Equation 28

one can obtain the values of \( \sqrt{Bn} \) for each laser excitation power. The IQE values could then be calculated using \( IQE = \frac{\left( \sqrt{Bn} \right)^2}{G} \). With an assumed \( B \) coefficient, the IQE values as a function of \( n \) could be obtained.

![Figure 61. Curve fitting results of the generation rate \( G \) as a function of integrated PL intensity \( I_{PL} \) for a c-plane LED active layer (with 6 nm InGaN DH) on sapphire. The red curve represents the fits obtained using Equation 25. (Fitting parameters: \( P_1 = 8.59437 \times 10^{23} \) and \( P_2 = 1.48422 \times 10^{22} \), goodness of fitting \( R^2 = 0.99863 \)).](image-url)
Figure 62. The internal quantum efficiencies of a c-plane LED active layer (with 6 nm InGaN DH) on sapphire, extracted from the excitation dependence of the PL intensity using a Ti-sapphire laser (370nm).
References


Vita

Xianfeng Ni
Date of Birth: December 4, 1979
Citizenship: P.R. China
Email: nixianfeng@gmail.com

Education:
Virginia Commonwealth University, Richmond, VA
Ph. D. in Electrical Engineering, Advisor: Hadis Morkoç

Zhejiang University, Hangzhou, P.R. China
M. S. in Semiconductor materials, June 2004

Zhejiang University, Hangzhou, P.R. China
B. S. in Materials science, July 2001

Work Experience:
08/2004 – 05/2010, Virginia Microelectronics Center, Richmond, VA
Research assistant (Advisor: Prof. Hadis Morkoç)
Epitaxy, fabrication and testing of polar and non-polar InGaN light-emitting diodes (LEDs)
MOCVD epitaxy of nonpolar, semipolar nitrides
Epitaxy and fabrication of nitride-based vertical-cavity surface emitting lasers (VCSELs)
LED device modeling with TCAD (Silvaco Atlas, APSYS)
MOCVD epitaxy of nitride high electron mobility transistors (HEMTs) structures, such as
AlGaN/GaN FETs, AlInN/GaN FETs.
PECVD deposition of dielectrics SiO₂, SiNx and related dielectric DBR

09/2001 – 06/2004, Materials department, Zhejiang Univ., Hangzhou, China
Research assistant
Deposition of SiGe and Si on Si.
Growth and characterization of GaN on Si substrate using MOCVD

Honors:
"Chinese Government Award for Outstanding Students Abroad" 2009
Member of American physical society
IEEE student member
Full member of Sigma Xi

Publications:
X. Ni, Q. Fan, R. Shimada, Ü. Özgür, and H. Morkoç, “Reduction of efficiency droop in InGaN


M. Wu, J. H. Leach, X. Ni, X. Li, J. Xie, Ü. Özgür, S. Dogan and H. Morkoç, "InAlN/GaN Heterostructure Field-Effect Transistors on Fe-doped Freestanding GaN Substrates", physica status solidi (a), (accepted)


Plus other coauthored journal papers and 6 oral conference presentations