QUANTUM EFFICIENCY ENHANCEMENT FOR GAN BASED LIGHT-EMITTING DIODES AND VERTICAL CAVITY SURFACE-EMITTING LASERS

Fan Zhang

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QUANTUM EFFICIENCY ENHANCEMENT FOR GAN BASED LIGHT-EMITTING DIODES AND VERTICAL CAVITY SURFACE-EMITTING LASERS

A research dissertation submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Electrical and Computer Engineering at Virginia Commonwealth University

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Abstract

QUANTUM EFFICIENCY ENHANCEMENT FOR GAN BASED LIGHT-EMITTING DIODES AND VERTICAL CAVITY SURFACE-EMITTING LASERS

By

Fan Zhang

A research dissertation submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Electrical and Computer Engineering at Virginia Commonwealth University

Virginia Commonwealth University, 2014

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This thesis explores the improvement of quantum efficiencies for InGaN/GaN heterostructures and their applications in light-emitting diodes (LEDs) and vertical cavity surface-emitting lasers (VCSELs). Different growth approaches and structural designs were investigated to identify and address the major factors limiting the efficiency. (1) Hot electron overflow and asymmetrical electron/hole injection were found to be the dominant reasons for efficiency degradation in nitride LEDs at high injection; (2) delta p-doped InGaN quantum barriers were employed to improve hole concentration inside the active region and therefore improve hole injection without sacrificing the layer quality; (3) InGaN active regions based on InGaN multiple double-heterostructures (DHs) were developed to understand the electron and hole recombination mechanisms and achieve high quantum efficiency and minimal efficiency droop at high injection; (4) the effect of stair-case electron injectors (SEIs) has been investigated with different active region designs and SEIs with optimized thickness greatly mitigated electron overflow without sacrificing material quality of the active regions. The active regions showing promising performance in LEDs were incorporated into VCSEL designs. Hybrid VCSEL structures with bottom semiconductor AlN/GaN and a top dielectric SiO₂/SiNₓ DBRs have been investigated, and quality factors as high as 1300 have been demonstrated. Finally, VCSEL structures with all dielectric DBRs have been realized by employing a novel ELO-GaN growth method that allowed integration of a high quality InGaN cavity active region with a dielectric bottom DBR without removal of the substrate while forming a current aperture through the ideally dislocation-free region. The full-cavity structures formed as such exhibited quality factors 500 across the wafer.
Chapter 1  Introduction

1.1 Motivation

In recent decade, III-nitride material has been extensively studied in application in optoelectronic devices in the blue spectrum to the ultra-violet (UV), including light emitting diodes (LEDs) and laser diodes (LDs), as well as applications in high frequency and power electronics. GaN materials are very promising in power devices due to the high breakdown field, high thermal conductivity, and high electron mobility (typically ~ 1600 cm$^2$/Vs at room temperature) in a typical AlGaN/GaN high electron mobility transistors (HEMTs) grown by metal-organic chemical vapor deposition (MOCVD). Moreover, tremendous improvements have been achieved in GaN-based LEDs. InGaN/GaN blue LED, which boost the LED market, was the last -- and most difficult -- advance required to create white LED light. With white LED light, companies are able to create smartphone and computer screens, as well as light bulbs that last longer and use less electricity than any bulb invented before. The Nobel Prize in Physics 2014 was awarded jointly to Dr. Isamu Akasaki, Dr. Hiroshi Amano and Dr. Shuji Nakamura for the invention of efficient blue LEDs which has enabled bright and energy-saving white light sources.

1.2 Development in GaN-based light-emitting diodes

GaN-based LEDs are used widely in various applications: liquid crystal display back lighting source, general lighting source, sensing, and communications. Unlike incandescent bulbs and fluorescent lamps, where most of the electricity is converted to
heat and only a small amount into light, GaN-based LEDs are capable of high efficiency in conversion electricity to light. A typical solid state LED features a chip of semiconductor heterostructures, i.e., InGaN/GaN, as a p-i-n junction. When electrons and holes are injected into the junction under forward bias, they either recombine radiatively to emit light or non-radiatively in the form of heating. GaN-based LEDs are more efficient with higher luminous flux (measured in lumen) per unit consumed power (measured in watt) compared to older light sources. The most recent record is just over 300 lumen/watt, which can be compared to 16 for regular light bulbs and close to 70 for fluorescent lamps, shown in Figure 1.1. As about one fourth of world electricity consumption is used for lighting purposes, the highly energy-efficient LEDs contribute to saving the resources consuming and significant reducing CO₂ and SO₂ gas pollution generated from burning of fuel for electricity.

![Figure 1.1](image.png)

**Figure 1.1**: LED lamps require less power to emit light than the older light sources.

Although the cost this emerging technology is an issue to be addresses, one can virtually certain that the cost of GaN related technologies will drop by orders of magnitude as this
technology matures. Yole Développement and EPIC announce their report dedicated to the LED industry in **Figure 1.2**.

![Globe LED market predicated by Yole Développement & EPIC](image)

**Figure 1.2: Globe LED market predicated by Yole Développement & EPIC**

As presented in **Figure 1.2**, the 2014 LED market size is estimated nearly $14.5 billion. Among the applications, currently about 14% of total sale accounted for mobile electronics, 21% for TV/monitors, and 48% for general lighting. It is expected that LEDs for general lighting will grow further in the next few years. To 2018, the percentage will be around 65% for the general light applications. Moreover, LEDs are constantly improved. Recent results from Soraa Inc. indicated that the peak light output densities of GaN-based LEDs exceeds 1000 W/cm² and their external quantum efficiencies (EQE) are maintained up to 56% at current density of 1000 A/cm² without current crowding.³⁴ However, challenges are also still prevailing in the process of LED market growth aside of high cost issues including LED performance at high power levels.
One of the lingering problems for high power GaN-based LEDs is the efficiency degradation, loss of efficiency at high electrical injection in LEDs beyond that which is expected from thermally activated processes. The EQE reaches its peak value at current densities as low as 50 A/cm² (in some cases, around 5 A/cm²) and gradually decreases with further increasing current injection\textsuperscript{5,6} Understanding the source of this degradation is critical and minimization is desirable for the implementation of the GaN-based LEDs. While the genesis of the efficiency degradation still remains a highly debatable topic, one obvious mechanism is the asymmetry in doping in wide bandgap semiconductors, such as GaN, wherein the hole injection falls behind that of electrons in the active region. The radiative efficiency cannot keep up with increasing carrier injection due to progressively lagging hole population in the active region. More importantly, Auger recombination and/or carrier overflow are considered as the most probably major cause for the efficiency degradation in InGaN LEDs.\textsuperscript{7,8,9,10} Further discussion about asymmetry electron/hole injection, Auger recombination and carrier overflow in InGaN/GaN active region and how to improve the quantum efficiency of LEDs will be present in Chapter 2 and Chapter 3.

1.3 Development in GaN-based vertical cavity surface-emitting lasers

The vertical-cavity surface emitting laser (VCSEL) has several advantages over the edge-emitting laser. Superiorities include the circular output beam, low beam divergence, high modulation band-width, single longitudinal mode, and convenient wafer level testing.\textsuperscript{11} Owing to the geometry of this class of laser, VCSEL is promising for optoelectronic devices for many applications, such as high-density optical storage system,
laser printing, and fiber-optic communications. Consequently, the VCSEL can combine relatively low manufacturing costs with great performance.

However, in contrast to the impressive progress of GaN-based LEDs and edge-emitting LDs, GaN-based VCSELs, which span 400 nm to 550 nm, still face significant challenges. To understand challenges of GaN-based VCSELs, one must first understand how this device operates. VCSEL is formed by sandwiched a thin active region between two parallel mirrors, and the cavity length is of a few microns – hundreds of times shorter than that of an edge emitting laser. The short cavity length enables high-speed direct modulation. However, the gain per round-trip pass through the cavity is far less than that for an edge-emitter. To compensate, cavity loss must be very small, and thus the reflectivity of the mirror, which is a distributed Bragg reflector (DBR), must be very high – it has to be nearly 99%. With a GaAs-based VCSEL, it is relatively easy to form such a high reflectivity with a stack of lattice-matched, quarter-wavelength thick alternating layers of GaAs/AlGaAs DBRs. Moreover, both GaAs and AlGaAs layers can be doped, thereby enabling electrical injection into the cavity. Replicating this approach with the III-Nitride growth has proved to be much hard. This had led several groups to introduce new types of device structures, which either combine an epitaxial DBR with a dielectric one, or employ dielectrics for both mirrors, which is shown in Figure 1.3.
Another consequence of the VCSEL’s short cavity length is the complex standing-wave profile of the electric field intensity. To optimize laser performance, it is essential to position the active region at the peak of this standing wave, while aligning lossy layers, such as heavily doped layers, at a standing wave null. Very precise control of the cavity length is needed to achieve this, shown in Figure 1.4. This is readily achievable in all-epitaxial structures, but more challenging in devices that feature double dielectric DBRs and are formed with substrate lift-off or thinning.
1.4 Scope of research

The research presented here concentrated primarily on GaN-based LED and VCSEL structure designs to enhance the quantum efficiency.

The dissertation is organized as follows: Chapter 2 is a discussion of efficiency droop issues persisted in the InGaN based LEDs. A review of the major causes will be included. Numerical simulation with Silvaco Altas will be included for theoretical prediction of our experiments. Our proposed mechanism will also be introduced and discussed.

Chapter 3 focuses on how to mitigate efficiency droop and improve quantum efficiency for GaN-based LEDs. Various structure designs will be presented and their effects on quantum efficiency will be discussed. Under this realm, the comparison of LED performance with/without delta p-doped barrier and with high or low height barrier to provide a understanding on efficiency limitation caused by poor hole transport. We will also present with multiple DH active region design and how the EQE was improved by
incorporating more thin DH in the active region. Finally, we presented that with optimized SEI for a given active region, the EQE can be further improved.

Chapter 4 focuses on quantum efficiency of GaN-based VCSELs. Various structure designs will be presented and their effects on quantum efficiency will be discussed. Under this realm, quality limitation of hybrid VCSEL will be discussed. We will also present the novel growth technique, fabrication process and measurement of VCSEL with both dielectric DBRs.

Finally, conclusions and suggestions for future work will be presented in Chapter 5.
Chapter 2 Efficiency droop investigations

2.1 Introduction to the efficiency droop

InGaN based light emitting diodes (LEDs) are becoming widely used for general lighting and displays with internal quantum efficiency (IQE) and optical extraction efficiency in high performance devices being in the range of 80\%.\textsuperscript{15} High power blue InGaN based LEDs are now able to produce over 250 lm/W efficiency with wall plug efficiencies, optical power divided by the electrical power, in excess of 60\% through improved quality and layer structural design, which is simply remarkable. However, LEDs suffer from the reduction of efficiency at high injection current levels. As shown in Figure 2.1, efficiency drops by about 50\% as current density increases from a few amperes per square centimeter to a few hundred amperes per square centimeter, which greatly affects the further application of LEDs. The DoE’s office of Energy Efficiency and Renewable Energy lists it as a top technical issue to be resolved in solid-state lighting.\textsuperscript{16}
To account for the efficiency droop, various models have been proposed, including current roll-over\(^{18}\), poor hole injection\(^{19}, 20, 21\), polarization field\(^{22}, 23\), Auger recombination\(^{24}\), and junction heating\(^{25}\). Weisbuch \textit{et al.} reported detection of Auger recombination in LEDs from the existence of Auger electrons by electron emission spectroscopy\(^{26}\), but it is unclear whether Auger recombination is the dominate cause of efficiency degradation or not. Moreover, the Auger loss in wide band-gap semiconductor as GaN is expected to be small, as verified by fully microscopic many body models\(^{27, 28}\). In addition, if Auger recombination was solely responsible for the efficiency degradation, GaN-based lasers, requiring high injection levels, would be undoubtedly prevented, which is not the case\(^{29}\). So far, the extensive experimental results suggest that the efficiency droop is related to the skewed carrier injection due to the disparity of hole and electron concentrations, large hole effective mass, and carrier overflow instead of the Auger recombination. As reported, Xie \textit{et al.} employed either \textit{p}-type doped InGaN
barriers or lightly $n$-type doped GaN electron injection layer just below the MQWs to achieve comparable levels of electron and hole injection and observed mitigation of the droop, which suggest poor hole transport and injection through the barrier being the responsible mechanism.\textsuperscript{30} Moreover, Ni et al. offered further supporting data for the electron overflow by investigating the efficiency droop in double heterostructure (DH) LEDs with different hot electron stopper or cooler designs both theoretically and experimentally.\textsuperscript{31}

Here we propose the electron overflow and lagged hole injection to be the dominant mechanism responsible for the efficiency degradation. The term “overflow electrons” refers to the electrons which escape the active region without participating in any recombination process, 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 investigate possible solutions to eliminate the electron overflow via numerical simulation in this chapter. The discussion of improve hole injection of LEDs will be discussed in Chapter 3.

2.2 Hot electron model

In previous work, Ni et al. evaluated non-equilibrium electrons, \textit{i.e.} hot electrons, inside in the InGaN/GaN LEDs.\textsuperscript{32} The injected electrons, which acquire additional kinetic energy equal to conduction band offset when entering active region, can traverse the active layer by ballistic or quasi-ballistic transport without recombination and recombine in the $p$-GaN region. We should note that the nonradiative recombination is prevalent in $p$-type GaN due to extremely long lifetime. Dr. Ni used first-order estimation of the hot electron effect and explained our experimental data with varying barrier height of the
EBL. The calculation assumes that the electrons obey the Fermi-Dirac distribution in the n-GaN layer 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 InGaN active region ($In_{0.15}Ga_{0.85}N$ active region is typical used for LEDs in our experiments) upon injection. These hot electrons would either undergo thermalization and lose their excess energy mainly through interaction with LO-phonons or avoid thermalization and escape the InGaN region as depicted in Figure 2.1.

**Figure 2.1:** 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 $E + \Delta E_c + qV(x)$. 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.

The calculations on the hot electron overflow took 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 one LO-phonon emission or absorption), and two scattering events (4
combinations of two scattering events involving LO-phonon emission and absorption). 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. The calculated contribution of electrons undergoing two scattering events to the overflow is less than 1% of the total injected electrons and can be neglected in our experimental device structures. Thus, based on the following three scattering events: (1) – no scattering, (2) – one phonon emission, (3) – one phonon absorption, the hot electron overflow can be estimated.

2.3 Stair-case injector (SEI) designs

In this section, we will concentrate on the designs of the SEI structures for optimum impact. Theoretically, 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, due to the growth technical limitations, thicker InGaN SEI layers have better chance to be relaxed and thus more threading dislocations could be generated and penetrated into the active region leading to the much increased non-radiative recombination. Therefore, growth related issues should also be taken into consideration when optimizing the SEI layer stack, which will be discussed in details later. The schematic structure is shown in Figure 2.2.
Figure 2.2: A schematic for the conduction band of a LED with a two-step layer SEI. ($\Delta E_c \geq 88\text{meV}$) After being injected into the SEI from the $n$-GaN region, some electrons will have ballistic and quasi-ballistic, while the others (experiencing two or more scattering events) are considered to be thermalized in the SEI.

A universal solution in the InGaN based LED structures in commercial companies to prevent electron overflow is to employ $p$-AlGaN electron blocking layer (EBL). However, it should be noted that the EBL impedes hole injection due to the generated valence band potential barrier between active region and $p$-GaN. Moreover, in the commercial $c$-plane GaN based LEDs, the AlGaN EBL is located on top of the InGaN barrier of the active region. The lattice mismatch between AlGaN and InGaN layers generates piezoelectric polarization field in addition to differential spontaneous polarization fields, which pull down the conduction band at the AlGaN/InGaN interface. As a result, the effective barrier height of the AlGaN EBL is a compromise, and the electron overflow is not effectively suppressed. InGaN SEI, inserted between $n$-GaN and InGaN active region, can reduce the electron overflow by gradually thermalizing down the hot electrons without blocking hole injection. It was demonstrated experimentally that with the insertion of SEI, the efficiency degradation of LEDs could be substantially reduced and
the AlGaN EBL, employed to reduce electrons overflow but hampers hole transport, could be safely replaced with the SEI.\textsuperscript{35} LED structures grown on non-polar $m$-plane bulk GaN with 6-nm DH active region show almost identical dependence of EQE on current density when they contain either a three-layer SEI only or both the SEI and an EBL, while the first-order calculations of the electron overflow at different applied forward voltage lead to a high overflow percentage from 60% to 90% for the LEDs without any SEI and EBL and this percentage can be significantly reduced down to 10-20% by inserting the SEI only. Furthermore, the first order calculation for the $c$-plane 6-nm DH LEDs reveals that the electron overflow for the LED with one-layer SEI (no EBL) saturates at \textasciitilde 18% once the applied voltage exceeds 6 V, while the overflow increases with the applied voltage from 0% to 50% when the voltage increases from 3 to 14 V for the LED with EBL if no SEI is incorporated. Therefore, most of the investigated LED structures in this thesis have incorporated SEI structures without employment of EBL unless indicated specifically.

It should be noted that the specifics of the SEI region would depend on the specifics of the active region, thicker active regions relaxing the design parameters. In this context, the electron overflow vs. injection current for a variety of single and multi In\textsubscript{0.15}Ga\textsubscript{0.85}N DH LEDs with two-layer SEIs of different thicknesses (with no electron blocking layer) has been calculated. For simplicity, the electrons are assumed to move in the direction normal to the hetero-interfaces only and the total electron overflow is obtained by summing the contributions from both ballistic and quasi-ballistic electrons for a given bias voltage. The injected current density vs. bias was obtained using a commercial
simulator, SILVACO ATLAS, with parameters appropriate for nitride materials as reported in Ref.35.

Figure 2.3: Electron overflow percentile in (a) single DH LEDs and (b) multi-DH LEDs with different SEI thicknesses and active region designs as a function of injected current density.

Figure 2.3 shows the schematic and the computed electron overflow percentiles for two types of LED structures (no EBL involved): (1) single DH LEDs with In$_{0.15}$Ga$_{0.85}$N active-region width varying from 3 to 9 nm and (2) multi-DH LEDs with different number of In$_{0.15}$Ga$_{0.85}$N active regions separated by 3 nm In$_{0.06}$Ga$_{0.94}$N barriers or GaN barriers. As evident from Figure 2.3 (a), the rate of the electron overflow reduces with increasing active region width. In the absence of an SEI, the overflow percentile for the single 3 nm DH LEDs reaches 68% at ~75 A/cm$^2$, while only 28% of electrons escape to the p-GaN in the 9 nm DH structure. At 150 A/cm$^2$ (injection current density beyond that used in general-lighting LEDs), the electron overflow reduces from 75% in 3 nm to 45% and 32% for the 6 nm and the 9 nm DH structures, respectively. These values are reduced
to 31%, 16%, and 10% for the 3 nm, the 6 nm, and the 9 nm DHs, respectively, when a 4+4 nm-thick two-layer SEI is employed and further to ~5% with a 20+20 nm thick SEI. It should be noted, however, that although the carrier overflow reduces with increasing DH thickness, the use of DH active regions with thickness beyond 6 nm may not be beneficial, partly because nonradiative recombination rate increases with increasing active region thickness beyond certain thickness due to material quality degradation. Furthermore, too wide active region would substantially reduce radiative recombination efficiency because of poor overlap of electron and hole wavefunctions in wide active regions. Actually it is promising to feature structures like multiple quantum wells (MQWs) or DHs with thin active layers but cumulatively sufficiently thick active regions. Figure 2.3 (b) illustrates that the thick cumulative active region naturally provides efficient electron cooling. Moreover, when the band offset between n-In$_{0.01}$Ga$_{0.99}$N underlying layer and barriers in the active region becomes larger than the LO phonon energy of 88 meV, the electrons are thermalized efficiently because the whole active region inclusive of the In$_{0.06}$Ga$_{0.94}$N barriers works naturally as an electron cooler. Owing to the relatively small conduction band discontinuity between the In$_{0.06}$Ga$_{0.94}$N barriers and the In$_{0.15}$Ga$_{0.85}$N active layers, the reduced barrier heights not simply contribute to the electron thermalization and lower the velocity of electrons entering the active regions, but also benefit hole transport in the active region. As evident from Figure 2.4 (b), the electron overflow percentile in the quad 3 nm DH without SEI or EBL reduces from 28% to 16% at 75 A/cm$^2$ when In$_{0.06}$Ga$_{0.94}$N is used as the barrier material instead of GaN and from 56% to 31% at 500 A/cm$^2$. 
2.4 Graded Electron Injector (GEI)

Graded electron injector (GEI) is using a stepwise InGaN electron cooling layer with increasing In composition gradually, compared with two-step staircase electron injector. The conditions imposed on heterojunction discontinuities in the case of SEI are eliminated entirely as the structure automatically cools electrons when they have sufficient potential energy to emit LO phonons. Moreover, electrons in the graded injector are continuously accelerated in the direction normal to the heterointerface due to the electric field and more efficient electron cooling is provided compared with SEI with the same total electron injector thickness, shown in Figure 2.4. Furthermore, as in the case of the SEI design, the total thickness along with the active layer would have to be considered in the realm of strain and strain relaxation with its ensuing potential defect generation.

![Figure 2.4: Band structure of single 3 nm DH with 40 nm-thick GEI and SEI under current injection 100 A/cm². GEI eliminates design requirements associated with the stepwise graded approach. Moreover, GEI provide more efficient electron cooling.](image-url)
cooling compared with two-step SEI with the same total electron injector thickness.

Figure 2.5: Electron overflow percentiles calculated for (a) single and (b) quad 3 nm DH LED structures with either SEI or GEI of various thicknesses.

**Figure 2.5** compares the electron overflow percentiles calculated for single and quad 3 nm DH LED structures with either SEI or GEI. It is clear that the elimination of band discontinuities in the GEI reduces the electron overflow considerably. For single 3 nm DH LEDs, the overflow at 100 A/cm$^2$ is reduced from 26 % with 4+4 nm thick SEI to 12 % with an 8 nm thick GEI. These values are reduced to 12 % and 10 % for 20+20 nm SEI and 40 nm GEI, respectively. For quad 3 nm LEDs, where the thicker active region naturally contributes to electron cooling, the overflow at 100 A/cm$^2$ is reduced from 10 % with 4+4 nm SEI to 7 % with an 8 nm thick GEI. The corresponding values are 5.5 % and 3.5 %, respectively, for quad 3 nm LEDs with 20+20 nm SEI and 40 nm GEI.
2.5 Electron leakage from active region

The electron overflow in forward biased LEDs must be delineated from the carriers’ recombination, including radiative and nonradiative recombination, by hot electron models and thus somewhat indirect. In order to have a clear picture of electron overflow, we measured electron leakage from the active region in reversed biased LEDs using the photodiode configuration with optical excitation, where electrons were traversing to the $n$-GaN of LEDs. The portion of the generated electron-hole pairs that participates in radiative recombination can be estimated by luminescence emanating from the junction. The contribution by the nonradiative component can be neglected as the layer quality nowadays is sufficiently good to justify this assumption. In the experiments we undertook, CW 325 nm HeCd excitation had to be used which is absorbed by the p-layer and the fraction of photons absorbed in the active region can only be determined if the minority carrier diffusion length is known.

2.5.1 Diffusion length measurement in p-GaN

First, we must determine the diffusion length by progressively thinning the p-layer, which are then used in the estimation of the photons that are absorbed in the active region in conjunction with the photodiode experiments. Here, we investigated diffusion lengths of the photo-generated carriers in $p$-type GaN along the $c$-direction, using photoluminescence (PL) spectroscopy, which do not require advanced nonlinear optical techniques or electrical contacts as for most of the abovementioned methods.
The samples used were c-plane 6 nm thick In$_{0.15}$Ga$_{0.85}$N double heterostructure (DH) active regions grown on a ~3.7 µm-thick n-type GaN template on sapphire in a vertical low-pressure metalorganic chemical vapor deposition (MOCVD) system. A 60 nm Si-doped ($2 \times 10^{18}$ cm$^{-3}$) In$_{0.01}$Ga$_{0.99}$N underlying layer was inserted just beneath the active region for improving the quality of the overgrown layers. The structures were completed with either Mg-doped (chemical doping concentration $\sim 10^{19}$ cm$^{-3}$) p-GaN layer of 500-nm thickness, shown in Figure 2.6. Hole concentration in p-GaN was determined to be $4 \times 10^{17}$ cm$^{-3}$ from Hall measurements on a separate calibration sample. To mitigate the effect of any Mg out-diffusion from p-GaN on the optical quality of the active region, a 20 nm thick In$_{0.01}$Ga$_{0.99}$N spacer layer was grown in between.

![Diagram](image)

**Figure 2.6:** Cross-sectional schematic of the InGaN-based DH samples investigated. The steps of different height are generated by ICP etching.

To achieve $p$-type layers with different thickness, selective area inductively coupled plasma (ICP) etching was used in multiple steps. ICP etching introduces surface damage
which modifies the PL intensity. To minimize this effect, a two-step etching procedure, a high power, i.e. physical etch step followed by a low power, i.e. chemical etch, was employed. This two-step etching process was found to help with the surface recovery so that the effect of etching on PL intensity could be neglected. Etched thickness of the top layer was determined after each etching step using a surface profiler. PL measurements were carried out on regions with different p-GaN thicknesses using He-Cd laser (325 nm wavelength) excitation with resulting photogenerated carrier concentrations \((n)\) from \(1.5\times10^{18}\) cm\(^{-3}\) to \(4.2\times10^{18}\) cm\(^{-3}\). At a given excitation power, the carrier density was determined from the generation rate at steady state, \(G = An + Bn^2 + Cn^3\), assuming that electron and hole capture cross-sections are equal, and \(n\) is much higher than the background carrier concentration, which is justified (background concentration \(\sim10^{17}\) cm\(^{-3}\)). The Shockley-Read-Hall, bimolecular, and Auger recombination coefficients of \(A = 10^7\) s\(^{-1}\), \(B = 5\times10^{-11}\) cm\(^3\)s\(^{-1}\), and \(C = 10^{-31}\) cm\(^6\)s\(^{-1}\), respectively, were used.\(^{36}\) The carrier generation rate, \(G\), was calculated using \(G = \left[P\alpha(1-R)\right]/(Sh\nu)\), where \(P\) is the excitation laser power incident on the sample, \(\alpha\) is the absorption coefficient at the excitation photon energy \(h\nu\), \(R\) is the Fresnel reflection coefficient for the sample/air surface, \(S\) is the laser spot size on the sample surface. Due to the large absorption coefficient \((1.1\times10^5\) cm\(^{-1}\))\(^{37}\) at the excitation wavelength, carrier photogeneration takes place near the surface region of the sample, and upon diffusion of carriers away from the surface the PL emanating from the active region, spacer layer, underlying InGaN and underlying n-GaN is measured. The thicker the top layer, the less intense the PL is from the underlying layers. The PL intensity of the etched region was compared with that from the corresponding un-etched reference region to minimize the effect of any variation across the sample. It
should be noted that as the top GaN layer is made thinner, more of the excitation light penetrates into the active region and the underlying layers and gets absorbed there. The resulting PL is then due to recombination of both the directly photogenerated carriers and those diffusing from the top GaN.

**Figure 2.7** shows the integrated PL intensity of the active region plotted as a function of the p-GaN layer thickness at 295 K and at 15 K, taking into account the absorption of incident laser power in the top p-GaN and In$_{0.01}$GaN spacer layer. With increasing p-GaN layer thickness, the normalized PL intensity exhibits an exponential decay of the form $e^{-x/L_{\text{diff}}}$, where $x$ is the p-GaN thickness and $L_{\text{diff}}$ is the diffusion length. The diffusion lengths in p-GaN extracted from the fits are 93±7 nm and 70±7 nm at 295 K and 15 K, respectively. The error in the diffusion length measurements by PL originates mainly from the thickness nonuniformity across the sample introduced by multiple ICP etching steps. A typical PL spectrum (region with 100 nm thick p-GaN) is shown in the inset of Fig. 2. As is often the case, it should be noted that due to high Mg doping, there is nearly no near band edge emission from the p-GaN layer.
Figure 2.7: Integrated PL intensities from underlying GaN, In$_{0.01}$GaN layer, and the active region at 15 K and 295 K as a function of p-GaN thickness. The lines are exponential fits to the data for the active region. Data at 295 K and 15 K are shifted vertically for clarity. Error bars apply to all respective points for a given thickness. The inset shows a representative PL spectrum for the region with 100 nm thick p-GaN at 295 K.

The decrease in diffusion length at low temperature in p-GaN is mainly due to the increased ionized impurity scattering which is dominant at low temperatures. As the thermal velocity of the carriers reduce, the effect of long-range Coulomb interactions on their motion increases. Moreover, radiative recombination rate increases with decreasing temperature, and consequently, shorter recombination lifetime reduces the diffusion length. This temperature dependence of diffusion length is also consistent with data available in literature.\textsuperscript{38}
2.5.2 Electron escape from active region

![Graph showing electron escape from active region]

Figure 2.8: Electron flow percentile out of the active region into the adjacent n-type semiconductor of an InGaN LED used as a photodiode with photogenerated electron density to be the parameter.

Figure 2.8 shows the percentile of electron leakage out of the active region vs. the bias for an LED containing a 6 nm-thick In$_{0.15}$Ga$_{0.85}$N active layer and a 10 nm-thick Al$_{0.20}$Ga$_{0.80}$N EBL. Naturally an increase with bias ensues because of the band bending due to the bias favoring electron escape from the active region. LEDs and photodiodes are dissimilar in that in the former one desire to have all the carriers to recombine in the active region and the recombination current to be the only current component. In the latter one desires to extract all the photogenerated carriers out of the absorption region which means that a well-designed LED would not make a very good photodetector but for the sake of the experiments discussed here, LEDs can be used in the photodiode realm. The photocurrent method can
be used to estimate the carrier overflow in LEDs by investigating structures with barriers of different height inserted on both sides of the active region.

### 2.6 Carrier overflow versus Auger recombination

Using the assumption that the recombination region is much thinner than the natural diffusion length of minority carriers for DH or MQW LEDs, the recombination rate equation under steady state is typically described as $A_n + B n^2 + C n^3 = J / q d$. The term $C$ represents Auger nonradiative coefficients. Therefore, in the absence of carrier spillover the IQE can be written as $\eta_{\text{int}} = \tau_{\text{eff}} / \tau_{\text{r}} = B n^2 / (An + Bn^2 + Cn^3)$. If it were possible to measure IQE vs. the injected current density $J$, one could obtain $A$, $B$, and $C$ coefficients through a third order polynomial fitting, albeit without a unique solution. The collected radiative power can also be measured vs. the optical excitation power from which $A$ and $C$ coefficients can be deduced for a given $B$ coefficient assuming that the extraction and collection efficiency remains the same for all excitation levels. Let us now turn our attention to calculating $A$ and $B$ coefficients first. Assuming a trap density of $N_t \approx 10^{16}$ cm$^{-3}$, a capture cross section of $\sigma = 10^{-15}$ cm$^2$ for this particular trap, and a thermal velocity of $v_{th} = 5 \times 10^6$ cm/s, which are reasonable, $A$ coefficient is found to be $A = 1/\tau_{\text{sr}} = \sigma v_{th} N_t = 5 \times 10^7$ s$^{-1}$, which is consistent with other report where this coefficient is extracted from a fitting of the light output dependence on the injection current density. For a detailed calculation of the $B$ coefficient, one can refer to Ref. 30, but in a simple sense, $B = G/n_i^2 \approx 4.1 \times 10^{-9}$ cm$^3$ s$^{-1}$ for GaN, where $G$ is the generation rate per unit volume and $n_i$ is the intrinsic carrier concentration. Doing so leads to a $B$ coefficient value for In$_{0.2}$Ga$_{0.8}$N of approximately $1.2 \times 10^{-10}$ cm$^3$ s$^{-1}$. These calculated $A$ and $B$ values
will serve as reference ranges for those extracted from fits to the curves of the light output vs. injection current density in order to reduce the error in values obtained from the polynomial fitting.

According to Ref. 41 and as shown in Figure 2.9, the calculated $C$ values for both LEDs are already out of the expected range of $1.4 \times 10^{-30}$ cm$^6$s$^{-1}$ to $2 \times 10^{-30}$ cm$^6$s$^{-1}$ predicted in Ref. 24.
Exp. data - Nichia LED

Fit

\[ A = 1 \times 10^7 \text{s}^{-1} \]
\[ B = 1.9 \times 10^{-10} \text{cm}^3 \text{s}^{-1} \]
\[ C = 3.2 \times 10^{-29} \text{cm}^6 \text{s}^{-1} \]

IQE

Current density (A/cm$^2$)

Lumileds

\[ A = 5.4 \times 10^7 \text{s}^{-1}, \ B = 2 \times 10^{-11} \text{cm}^3 \text{s}^{-1}, \ C = 2 \times 10^{-30} \text{cm}^6 \text{s}^{-1} \]

Fit (fixed \( B = 1.9 \times 10^{-10} \text{cm}^3 \text{s}^{-1} \))

\[ A = 1.75 \times 10^8 \text{s}^{-1} \]
\[ C = 5.5 \times 10^{-29} \text{cm}^6 \text{s}^{-1} \]
Returning to more conventional experiments, there are also other observations to identify effect of Auger recombination in carrier recombination. Binder *et al.*\(^4\) investigated two color structures wherein when the longer wavelength layer (green) is resonantly excited, the shorter wavelength (UV) region emits light, which was attributed to hot electrons created by the Auger process in the longer wavelength layer (green) traversing to the shorter wavelength one (UV) followed by radiative recombination. This report, however, is void of quantum well (QW) thicknesses and excitation power density levels used. Moreover, Hader *et al.*,\(^4\) using a fully microscopic model to emulate this particular experiment, argue that resonant laser tuned to create carriers only in the green region does also create carriers in the UV region even in the absence of any Auger process. The reason provided is that the polarization excited by the spectrally narrow optical pulse dephases in few tens of femtoseconds due to electron-electron and electron-phonon scattering. In this scenario, the free carriers are generated through a coupling between the optical pulse and the associated material polarization causing the total spectral width of the excitation to be dominated by dephasing time of the polarization, not by that of the optical pulse. The resulting spectral width is argued to be sufficiently wide to excite the UV QW as well. Due to the very small coupling between the states in the green region and those of the UV region, the carriers generated in the UV region do not relax to states in the green region, which paves the way for UV emission. In our own experiments dealing with two color systems, the resonant excitation of the blue QW region using a frequency doubled Ti:Sapphire laser of ~ 100 fs pulsewidth and 80 MHz...
repetition rate did not result in any detectable emission from the UV QWs (400 nm) up to the maximum laser excitation density that can be applied in our lab which corresponds to a carrier density of $10^{19}$ cm$^{-3}$, much larger than the injection densities employed in LEDs, as can be seen in Figure 2.10. It should also be noted that this carrier density is estimated using the steady-state approximation, which certainly is not fully valid for the pulsed excitation conditions used, and therefore, represents an average value and only a lower limit of the instant carrier density within the active region.

![Plot of PL intensity vs wavelength with a peak at 460 nm and a gap at 400 nm.](image)

**Figure 2.10:** With 425 nm excitation at 15 K, only the PL peak from low energy blue QW (460 nm) can be observed at an excitation density corresponding to $10^{19}$ cm$^{-3}$ photo-generated carrier concentration. Note the lack of emission from the larger bandgap UV QW (400nm). The spectral range near the excitation laser line is blocked.

When the two-color LED mentioned above was excited using a 1 kHz repetition rate 130 fs pulsewidth optical parametric amplifier (425 nm wavelength) providing much higher photon fluences allowing clear observation of two-photon absorption related emission from all layers, still no evidence of Auger recombination related transfer of hot electrons from the blue QW to the UV QW was observed. Error! Reference source not found.
2.11 (a) shows the PL intensities for the UV and blue QWs as well as the underlying GaN layers as a function of the excitation density. Under the excitation conditions used the PL intensities from the UV and blue QWs are proportional to $Bn_{UV}^2$ and $Bn_{blue}^2$, where $n_{UV}$ and $n_{blue}$ are the electron densities in the UV and blue QWs, respectively. If Auger recombination were effective in the blue QWs, their generation rate ($Cn_{blue}^3$) would be equal to the generation rate in the UV QWs ($Bn_{UV}^2$ under steady-state) following hot electron transport from the blue QWs under the assumption that Auger recombination in the UV QWs and the SRH recombination overall at 15 K are negligible. This would then imply that the intensity from the UV QWs should exhibit cubic dependence on the square root of the intensity from the blue QWs. However, the observed dependence above the threshold for the UV QW emission is to the 6th power as shown in Figure 2.11 (b).

Figure 2.11: (a) Integrated PL intensities for the UV and blue QWs and the GaN layer in the two-color LED with 425 nm excitation at 15 K. (b) PL Intensity from the UV QWs versus the square root of the PL intensity from the blue QWs, which is proportional to the electron density in the blue QWs, $n_{blue}$.
The observed quadratic dependence of the UV QW PL on excitation density in Figure 2.11 (a) suggests that the UV emission is due to generation of carriers in the UV QWs by two-photon absorption. The same quadratic dependence is observed for the PL from the underlying GaN layer. It can, therefore, be concluded that the Auger recombination is not effective at carrier densities that correspond to the threshold for UV emission, beyond which significant two-photon absorption occurs. The average carrier density estimated for the threshold excitation density value of 100 $\mu$J/cm$^2$ is $2 \times 10^{19}$ cm$^{-3}$, which, as indicated above, represents only a lower limit due to the femtosecond pulsed excitation used.
Chapter 3  Quantum efficiency enhancement in GaN-based LEDs

3.1 Effects of quantum barrier: height and thickness

3.1.1 Motivation
We have undertaken a series of theoretical investigations to unveil the root cause of the dominant efficiency degradation mechanism: hot electron overflow and lagged hole injection compared with electron injection. In order to retain the quantum efficiency at high injection, one approach would increase of the number of quantum wells (QWs) in the active region. However, due to the poor hole transport in InGaN LEDs, light emission is attributed mainly to the QWs closest to the $p$-GaN layer in typical $c$-plane GaN-based LEDs. Another approach is to utilize double heterostructure (DH) active regions for uniform carrier spreading across the active region. In this realm, different barrier height (either In$_{0.01}$GaN or In$_{0.06}$GaN barriers) and thickness (3 nm and 12 nm) applied on MQWs were investigated.

3.1.2 Experimental procedures
The $c$-plane InGaN LED structures, emitting at ~420 nm, were grown on ~3.7 µm-thick $n$-type GaN templates on sapphire in a vertical low-pressure metalorganic chemical vapor deposition (MOCVD) system. The structures feature various active regions such as variants of MQWs and DHs, with two-step SEI (two 4-nm InGaN layers with step-increased Indium composition of 4% and 8%) for optimization of internal and external quantum efficiencies. The SEI steps have conduction band potential energy drop equal to or more than one LO-phonon energy (92 meV for GaN) and enhance electron thermalization through LO-phonon emission. The MQWs active regions are with
different barrier height (either In$_{0.01}$Ga$_{0.99}$N or In$_{0.06}$Ga$_{0.94}$N barriers) and thickness (3 nm and 12 nm). All the structures contained a 60-nm Si-doped ($2 \times 10^{18} \text{ cm}^{-3}$) In$_{0.01}$Ga$_{0.99}$N underlying layer below the active region and SEI for quality improvement. The LED structures were completed with 100 nm-thick Mg-doped $p$-GaN layers having $4 \times 10^{17} \text{ cm}^{-3}$ hole density, determined by Hall measurements on a separate calibration sample. For devices, square mesa patterns (400x400 $\mu$m$^2$) were formed by conventional lithography and chlorine based Inductively Coupled Plasma (ICP) 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/5nm Ni/Au electrodes served as the semi-transparent $p$-contact, with 40/50-nm Ni/Au electrodes deposited for $p$-contact pads. The fabricated device is shown in Figure 3.1.
3.1.3 Results and discussion

The relative EQE is measured on-wafer pulsed EL measurements (0.1 % duty cycle, 1 kHz) carried out for all the investigated LEDs with no effort having been made to enhance light extraction. Figure 1 (a) shows the schematic for the LED structures with coupled-MQW In_{0.01}Ga_{0.99}N high barrier (HB). Figure 1(b) shows the relative EQE values for the MQW-LED structures. Among them, the relative EQE for the low barrier (LB) MQWs is always higher than MQWs with high barriers (30% in coupled-MQWs series and 15% in uncoupled-MQWs series). It is well known that the hole transport in GaN is compromised due to the large hole effective mass and ensuing low hole mobility. The lower In_{0.06}GaN barrier height in the MQW active regions would help the hole transport and quantum tunneling ascribed to the lowered barrier and thus reduce the electron overflow induced efficiency degradation at high injection levels as the probability of recombination in the active region would increase with more holes present.

Figure 3.1: Device schematic of fabricated LED with contacts.
Figure 3.2: (a) Schematic conduction band profile of coupled MQWs with HB with flat band for simplicity. (b) Relative EQE of MQWs LEDs as a function of pulsed injection current density (0.1 % duty cycle and 1 kHz frequency).

Comparing the coupled-MQW-LB to the uncoupled-MQW-LB-LED, the peak EQE for the former is 25% higher than that for the latter. We think that it is due to the relatively enhanced hole transport through the thin and low InGaN barrier in MQW active regions and resulting more efficient recombination of electrons and holes barring the complication caused by the induced field. It is clear that the coupled MQWs with 3 nm barriers would help provide a more uniform hole population among the 6-period quantum wells than the uncoupled MQWs. In the uncoupled (12 nm barrier) MQW-LEDs, the hole concentration dominates in the QW near the p-side, while for the coupled (3 nm barrier) MQW-LEDs holes can be more uniformly distributed across all the QWs.
Therefore, all the 6 wells are more likely to participate nearly fully in the recombination process in coupled MQW-LEDs, which thereby reduces the excess electron density and thus electron leakage. As for the efficiency degradation ratio (taken as the EQE value at 600 A/cm² relative to the maximum EQE), the LEDs with coupled MQW active layers show smaller degradation ratios compared with the uncoupled MQW counterparts. A coupled-MQW-LB-LED investigated shows negligible efficiency degradation while the uncoupled-MQW-LB-LED counterpart shows 25% degradation, which can be attributed to the enhanced hole distribution through thinner barriers.

3.2 Quantum efficiency for multi-DH LEDs

3.2.1 Motivation
As GaN-based LED technology continues to develop and mature, high brightness LEDs retaining high quantum efficiencies at high injection levels (>100 A/cm²) have become even more desirable to replace the prevailing incandescent lamps and fluorescent tubes in general lighting. However, the quantum efficiency of typical InGaN multi quantum well (MQW) LEDs peaks at current densities even as low as ~ 10 A/cm², and drops with increasing injection by a factor of as much as 2 in some reported cases. Although debates still persist on the origins of “efficiency droop”, carrier overflow has been reported to be the substantial component. In order to avoid carrier overflow and increase the light output, LEDs must employ thick double heterostructure (DH) or MQW active regions.

In conventional InGaN/GaN MQWs, normally thick (>10 nm) GaN barriers were employed to ensure active layer quality. However, electron and hole mainly recombine in the QWs closest to p-GaN due to the poor hole transport. We have already exhibited
with employment of thin low height In$_{0.06}$GaN barriers, hole population in the active layer was improved, resulting higher quantum efficiency and suppressed efficiency droop at high injection. DH active regions on the other hand can ensure more uniform hole spreading across the active region due to the absence of barriers and consequently have paved the way for negligible drop in quantum efficiencies beyond current densities of $\sim$150 A/cm$^2$.\textsuperscript{49} Moreover, DH LEDs possess bulk-like 3D density of states (DOS), and therefore, can accommodate more carriers than thin QWs having constant 2D DOS. However, among the ramifications of DHs are the degradation of InGaN structural quality with increasing thickness and separation of electron and hole wavefunctions due to the polarization field in the $c$-plane variety.\textsuperscript{50,51} Therefore, keeping the DH layer thin (3 nm) but stacking multiple of them separated by thin and low barriers (for ameliorating hole transport) in the active regions could be a promising approach to maintain high material quality and overcome the efficiency loss at high driving currents. In this work, we demonstrate that by using 3nm-thick multi-DH active layers separated by 3 nm-thick low-energy In$_{0.06}$Ga$_{0.94}$N barriers the electroluminescence (EL) is enhanced dramatically, in proportion with the number of DH layers up to 4 without discernible efficiency loss at high injection levels. Moreover, under resonant optical excitation, emission intensities at 10 K increase linearly with excitation power, indicating nearly unity quantum efficiency, and scale with the active region thickness for a given excitation density. We have proved that, with increasing number of active region DH layers, carrier overflow is unequivocally shown to reduce significantly. Active layers quality degrades beyond 6 DH layers due to the defects generation in the strain accumulation.
Among the ramifications of DH are the loss of InGaN quality and the increased band bending due to the polarization field. In spite of this, we have demonstrated that with increasing DH active layer thickness from 3 nm to 6 nm the relative EQE increased considerably with a given SEI design. The relative EQE was enhanced substantially in dual 3 nm DH and dual 6 nm DH LEDs, separated by a 3 nm-thick In$_{0.06}$Ga$_{0.94}$N barrier. Incorporating more DH active regions of the same thickness, separated by thin and low InGaN barriers, results in enhanced emission intensity without any discernible degradation of the active region quality unlike that observed in thicker single DH layers due to strain relaxation with increasing InGaN thickness.

### 3.2.2 Experimental procedures

The $c$-plane multi-DH InGaN LED structures, emitting at ~425 nm, were grown on ~5 µm-thick $n$-type GaN templates on sapphire substrates in a vertical low-pressure metalorganic chemical vapor deposition (MOCVD) system. The GaN templates employed an $in~situ$ SiN$_x$ nanonetwork to reduce the dislocation density down to mid-$10^8$ cm$^{-3}$. The active regions contained single or multiple In$_{0.15}$Ga$_{0.85}$N DH active layers separated by 3 nm In$_{0.06}$Ga$_{0.94}$N barriers, shown in Table 3-1. All the structures incorporate a staircase electron injector (SEI) for efficient thermalization of hot electrons prior to injection into the active region and a 60-nm Si-doped ($2 \times 10^{18}$ cm$^{-3}$) In$_{0.01}$Ga$_{0.99}$N underlying layer for improving the quality of overgrown layers. The SEI consists of two 4 nm InGaN layers with step-increased In compositions of 4% and 8%, inserted in the given order below the active region. The LED structures were completed with 100 nm-
thick Mg-doped $p$-GaN layers having $6 \times 10^{17}$ cm$^{-3}$ hole density, as determined by Hall measurements on a separate calibration sample.

**Table 3-1 LED structure of multiple DHs with sole SEI**

<table>
<thead>
<tr>
<th>LED</th>
<th>SEI</th>
<th>Active layers</th>
</tr>
</thead>
<tbody>
<tr>
<td>Single</td>
<td>4+4 nm</td>
<td>3 nm</td>
</tr>
<tr>
<td>Dual</td>
<td>4+4 nm</td>
<td>2x3 nm</td>
</tr>
<tr>
<td>Quad</td>
<td>4+4 nm</td>
<td>4x3 nm</td>
</tr>
<tr>
<td>Hexa</td>
<td>4+4 nm</td>
<td>6x3 nm</td>
</tr>
<tr>
<td>Octa</td>
<td>4+4 nm</td>
<td>8x3 nm</td>
</tr>
<tr>
<td>Single 6 nm</td>
<td>4+4 nm</td>
<td>6 nm</td>
</tr>
<tr>
<td>Dual 6 nm</td>
<td>4+4 nm</td>
<td>2x6 nm</td>
</tr>
<tr>
<td>Single 9 nm</td>
<td>4+4 nm</td>
<td>9 nm</td>
</tr>
<tr>
<td>Single 11 nm</td>
<td>4+4 nm</td>
<td>11 nm</td>
</tr>
</tbody>
</table>
3.2.3 Results and discussions

![Graph showing integrated PL intensity as a function of excitation power density at 10 K and 295 K.](image)

**Figure 3.3:** Integrated PL intensity as a function of excitation power density at (a) 10 K and (b) 295 K; gray solid lines indicate slope of 1 and the inset of (b) displays the PL-IQE vs. the number of 3 nm DHs in the active region; (c) PL efficiencies of multi-3 nm DHs vs. excitation power density at room temperature.

One common procedure to evaluate the IQE involves excitation-dependent photoluminescence (PL) measurements and comparison of the PL intensities at low and room temperatures by assuming 100% IQE at low temperature though it might not be the case.\(^{53,54}\) Excitation power dependent resonant PL measurements were performed at both 10 K and 295 K using 385 nm excitation from a frequency doubled Ti:Sapphire laser ensuring photo-generation of carriers only in the LED active regions. The highest excitation density used corresponds to an average carrier concentration of \(\sim 10^{18} \text{ cm}^{-3}\) in the single DH LED structure. As the collected PL intensity is proportional to excitation intensity, \(L_{\text{PL}} \propto I_{\text{exc}}^m\), the linear dependence \((m \approx 1)\) for all structures at 10 K in **Figure 3.3** (a) indicate that the radiative recombination dominates, \(i.e. \tau_{\text{Rad}} \ll \tau_{\text{nonRad}}\) where \(\tau_{\text{Rad}}\) and \(\tau_{\text{nonRad}}\) are the radiative and nonradiative lifetimes, respectively. It is therefore
reasonable to assume that the quantum efficiencies are nearly one at 10 K for all DH LEDs, omitting the negligibly slight deviations. The room temperature data shown in Figure 3.3 (b), however, shows superlinear dependence (m ≈ 1.4 - 1.95) for low excitations, which is attributed to the notable impact of nonradiative recombination (m = 2 in case of constant $\tau_{\text{nonRad}}$). As the excitation density is increased, the slope gradually approaches to $m = 1$ ($I_{\text{PL}} \propto I_{\text{exc}}$). The gradually decreasing slope in the intermediate excitation regime indicates strong competition between nonradiative and radiative processes and can be attributed to decreasing radiative lifetime, beneficial, for moderate injections. Another process to be kept in mind is that with further increase of excitation, saturation of localized states and delocalization of carriers (particularly holes, as the electron density in the wells is in mid-10$^{17}$ cm$^{-3}$) may allow access to additional nonradiative centers and result in enhanced recombination rate with respect to low excitation.$^{55,56}$ Moreover, Coulomb screening of the quantum confined Stark effect (QCSE) with increasing excitation leads to an increased interband recombination rate $1/\tau_{\text{Rad}}(N) \propto BN$, where B is the bimolecular recombination coefficient and $N$ is injected carrier density. The IQE values deduced from ratio of PL intensities at 300 K and 10 K at the highest excitation density employed are shown in the inset of Figure 3.3 (b). The quad 3 nm DH LED exhibits an IQE, so determined, of ~46% whereas increasing the number of 3 nm DH active regions to 6 and 8 lowers the IQE to 36% and 16% respectively, indicative of active region degradation with increasing overall thickness due to plausibly strain relaxation and increased interface roughness. This degradation is evident also from the room temperature PL efficiencies, defined as the collected integrated PL intensity normalized to the incident laser power, shown in Error!
Notably, the PL efficiencies nearly scale with the number of DH layers up to 6 due to increased absorption and emitting volume, showing ~2, 4 and 6.5 fold increase for dual, quad and hexa DHs compared to single DH at an excitation density of 1.5 kW/cm$^2$, but no further improvement for the octa DH LED which is most likely a manifestation of material degradation.

Figure 3.4: (a) The integrated EL intensity dependence on current density (the grey-solid line indicates slope of 1), (b) Relative EQE of multi-3 nm DHs vs. injected carrier density.

To study the impact of carrier overflow and other carrier transport features, we measured EL efficiencies on-wafer (unpackaged) with light output collected primarily normal to the sample surface by an optical fiber. The integrated EL intensities vs. injection current are shown in Figure 3.4 (a). The integrated EL intensity, $L_{EL}$, can be described by a power dependence on the injection current density as $L_{EL} \propto J^m$, where the power index $m$, as in the case of optical excitation, reflects an effective rate of recombination processes within a given range of current densities.\textsuperscript{57} The superlinear
growth of EL intensity ($m \sim 1.4$ for single, $\sim 1.3$ for dual and quad, and $\sim 1.6$ for hexa and octa DH LEDs) at low current densities is attributed to nonradiative recombination. Smaller $m$ values suggest lower density of nonradiative recombination centers in single, dual and quad DH LEDs compared to hexa and octa DH LEDs. The EL intensity changes nearly linearly at high current levels; therefore, relative EQE tends to be constant.

As presented in Figure 3.4 (b), the relative EQE for the multi-DH structures with up to 4 DH layers increases rapidly with current injection and reaches its maximum at $\sim 35\text{-}40$ A/cm$^2$. Compared to the single 3 nm DH LED, the peak EL efficiencies for dual and quad DH LEDs are higher by 1.6 and 3.5 times, respectively. Unlike in the case of optical injection, this significant improvement on EL efficiency cannot simply be explained by increased emitting volume as for a given current density overall carrier concentration is the same. Therefore, the data unequivocally indicates that increasing the number of 3 nm DH layers (from 1 to 6) decreases the overflow of injected carriers considerably (i.e. more of the injected carriers are captured by the active region), while further increase in number of DH layers (8) aggravates by introducing more nonradiative recombination centers due to degradation of the active region quality.

It is important to note that although the 4+4 nm SEI design used here has been shown to be an effective replacement for the Al$_{0.15}$Ga$_{0.85}$N electron blocking layer for reducing electron overflow in structures with multiple QWs and wider DHs (6 and 9 nm),$^{58}$ it must be optimized for a given active layer to fully prevent electron overflow. Based on the simulation results in Figure 2.3, the percentage of electrons captured by and recombine in the active region is increased to 76% in quad DH LED compared to 48% and 60% for single and dual 3 nm DH LEDs, respectively, at a current density of $\sim 500$ A/cm$^2$. LED
structures with optimized SEI layers should greatly reduce the carrier overflow if not eliminated while maintaining the active region quality. For optimum SEI layer design, which depends on the overall active region design, the resulting maximum EL efficiencies for single and quad 3 nm DH LEDs should be similar.

As observed in Figure 3.4, the relative EQE of hexa 3 nm DH LED is only slightly larger than that of the quad 3 nm DH, which suggests that the injected carriers are mostly consumed in the first four DH layers close to $p$-GaN due to limited hole transport for the achieved hole concentration and/or the active region quality may have slightly degraded with increased overall thickness. Further increase in the number of DH layers to 8 lowered the EL efficiency by ~20%, indicative of a clear indication of the active layer quality degradation, which is also confirmed by PL measurements conducted at 10 K and 295 K in Figure 3.3.

![Figure 3.5: Relative EQE of DH LEDs as a function of pulsed injection current density (0.1 % duty cycle and 1 kHz frequency).](image-url)
The relative EQE values for the LED structures with various DH thicknesses are shown in Figure 3.5. Single 3 nm DH LED shows around 25% of peak EQE value compared with that of the single 6 nm DH LED, indicative of severe electron overflow due to its thin active region. Dual 3 nm DH LED shows doubled EQE value compared with single 3 nm DH LED. The single 6 nm DH LED structure shows the maximum relative EQE values at current density ~41 A/cm², slightly higher than that for 31 A/cm² in 3 nm DH LED, indicative of a slower rate of increase with injection for the 6 nm DH LED. The 9 nm DH LED shows a slow rise of EQE with increasing current injection and reaches its maximum EQE at the injection current density as high as 220 A/cm². The slow initial EQE rise in the 9 nm DH is likely caused by both defects and slow increase of $B_{eff}$ coefficient with injection current, which will be discussed later. The relatively low maximum EQE can be ascribed to: (1) low value of radiative recombination coefficient because of wide separation of electron and hole wavefunctions, and (2) possible onset of strain relaxation giving rise to enhanced nonradiative recombination. Increase in DH active region width to 11 nm results in the further decrease of the maximum EQE and even slower EQE rise with injection current density. The 9 nm DH LED shows the highest EQE among the single DH LEDs studied at injection current densities above ~180 A/cm² with small efficiency degradation (~10% at 500 A/cm² with respect to the maximum). The dual 6 nm DH shows 20% higher peak EQE than that obtained from the single 9 nm DH and much faster increasing rate of EQE with current injection. At 200 A/cm², the relative EQEs retain 88% of the peak value for the dual 6 nm DH LEDs.

In conclusion, multiple DH structures have been demonstrated to enhance the quantum efficiency of GaN-based LEDs at high injection levels. We showed that multi-3 nm DH
active layers (having 3D-like DOS) represent an effective avenue to improve quantum efficiency with a given SEI design. Excitation dependent PL results indicate that PL efficiency is nearly proportional to the number of multi-3 nm DH layers up to 6 at room temperature, suggesting the same quantum efficiency for each DH active layer. Similarly, EL efficiency is also shown to increase with the number of DH active layers up to 4, due to reduced electron overflow, and the hexa DH LED shows ~20 % higher EL efficiency than that of quad DH LED at high injection. Increasing single DH thickness from 3 nm to 6 nm resulted in an increase in peak EQE. Further increase of the DH active region thickness to 9 nm improved EQE only at very high injection levels while 11 nm thick DH showed significantly lower EQE due to active region quality degradation. Therefore, among the efforts to enhance the quantum efficiency at elevated injection levels, multi-DH layer designs with appropriate electron injectors can constitute a viable alternative approach to achieve high efficiency and high power LEDs.

3.3 Optimization of stair-case electron injector

3.3.1 Motivation

We have undertaken a series of investigations to unveil the root cause of the efficiency degradation mechanism. Experiments conducted on multi-DH LEDs on c-plane GaN templates indicated that hot electron overflow is the dominant mechanisms in the EL efficiency loss of LEDs at high current level. This clue shows that hot electrons, either ballistic electrons or quasi-ballistic electrons, escape from the active region and recombine with the holes in p-GaN layer where the recombination is predominantly
nonradiative. In this section, LEDs with various active regions have been designed. Some of them incorporated single thicker active layer or multi-thin active layers with reduced barrier height favoring hole transport. The structures were employed with varied thickness SEI only (no EBL involved) to have a clear picture of SEI effects on electron cooling in LEDs.

As discussed in last section, the EL efficiency of quad 3 nm DHs shows about 3.5 times higher than that of single 3 nm DH. We attributed this to the more severe electron overflow in single 3 nm DH. By incorporating more 3 nm DHs, the active layer thickness is increased and thus electrons can be captured more readily in quad 3 nm DHs. First order calculation also indicated that only 48% of electrons can be efficiently injected in to the single 3 nm active region and recombine radiatively. As such, it is desirable to reduce the electron overflow by optimizing SEI structures. Here, we increased the two-step SEI thickness without modifying the In composition inside SEIs in order to cool down electrons sufficiently by keeping the total SEI thickness up to 60 nm. Table 3-2 shows the details of studied LED structures.

Table 3-2 LED structure with various active region designs and SEI thickness

<table>
<thead>
<tr>
<th>LED</th>
<th>SEI thickness (nm)</th>
<th>Active region (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LED-A</td>
<td>4+4</td>
<td>3</td>
</tr>
<tr>
<td>LED-B</td>
<td>20+20</td>
<td>3</td>
</tr>
<tr>
<td>LED-C</td>
<td>30+30</td>
<td>3</td>
</tr>
<tr>
<td>LED-D</td>
<td>4+4</td>
<td>Quad 3</td>
</tr>
<tr>
<td>LED-E</td>
<td>20+20</td>
<td>Quad 3</td>
</tr>
<tr>
<td>LED-F</td>
<td>30+30</td>
<td>Quad 3</td>
</tr>
<tr>
<td>LED-G</td>
<td>4+4</td>
<td>6</td>
</tr>
<tr>
<td>LED-H</td>
<td>20+20</td>
<td>6</td>
</tr>
</tbody>
</table>

3.3.2 Results and discussions
Before discussing the electron overflow in LED structures of different design, let us evaluate the effect of SEI on the optical quality of the LED active regions. For this purpose excitation density dependence of the resonant photoluminescence (PL) intensity was measured at room temperature to gain insight into the optical quality of these DH LEDs with varying SEI thickness, noting that in resonant excitation carriers are cool already. A frequency-doubled Ti:Sapphire laser (385 nm wavelength) was used with the maximum excitation density corresponding to an average carrier concentration of $\sim 10^{18}$ cm$^{-3}$ in the single DH LED. As shown in Figure 3.6 (a), PL intensity has a linear dependence on the excitation power for all single and quad samples at 15 K, which indicates that radiative recombination is dominant within the entire excitation density range employed. Moreover, the PL intensity scales nearly with the number of 3 nm DH layers in the active region: PL intensity for quad DH LEDs is $\sim 4$ times that of single DH LEDs. No change was observed in the emission wavelength with increasing SEI thickness for any of the LEDs, suggesting that there is no discernible difference in strain among the structures investigated. The PL intensities from the 6 nm DH LED structures are between those for single 3 nm DH and quad 3 nm DH LEDs.
Figure 3.6: The integrated PL intensity dependence on optical excitation density at (a) 15 K and (b) 295 K for single and quad 3 nm DH LEDs with varied SEI thickness.

Room temperature measurements [Figure 3.6 (b)] for the quad 3 nm DH LEDs show that at low injection the slope of PL intensity dependence on excitation density decreases with increasing SEI thickness, indicating reduced nonradiative recombination.
At a very low excitation density of 0.001 kW/cm$^2$, increasing the SEI thickness from 4+4 nm to 20+20 nm improves the PL intensity by nearly an order of magnitude, while no further improvement is apparent with the 30+30 nm-thick SEI. As the excitation density is increased, the slope gradually approaches 1, indicating a strong competition between nonradiative and radiative processes, with the latter becoming more prominent with increasing injection. The improvement observed at low excitation densities with increased SEI thickness suggests improvement of the active region material quality manifested as a reduction of nonradiative recombination centers although particulars of the genesis of this improvement is unclear at this instant. Similar trends are observed for the single 3 nm DH LEDs at room temperature except lower PL intensities due to their smaller active region thickness. Significant improvement is apparent in PL efficiency at low excitation for the single 3 nm DH LED with increasing SEI thickness from 4+4 nm to 20+20 nm. However, single 3 nm DH LED with 30+30 nm SEI exhibits the lowest PL efficiency at low excitation densities but the highest intensity at the highest excitation density employed. This is indicative of the presence of low density of certain type of nonradiative centers that can be saturated faster with increasing excitation compared to those in other LED structures. They may possibly originate from strain driven diffusion of Mg from the $p$-GaN layer, the detrimental effect of which will be much weaker in the quad LEDs, and/or generation of threading dislocations and related point defects within the active region resulting from partial relaxation due to accumulation of strain energy with increasing SEI thickness. Note that the detrimental effects of dislocations and point defects acting as nonradiative centers can be stronger in the single DH LEDs, which is consistent with the observations by Armstrong et al., who reported reduction in density...
of deep centers towards the LED surface, i.e. with increasing active region thickness, within multi-QW or multi-DH LEDs. This unusual behavior for the single DH LED with 30+30 nm SEI is still under investigation; however, it does not affect the premise of this paper as of particular importance is the LED operation at high excitation densities, where the single 3 nm DH LEDs with 20+20 nm and 30+30 nm SEIs have comparable performance. Moreover, it should be noted that the higher efficiencies (lower slopes) at low excitation densities for the quad DH LEDs compared to the single DH LEDs result in part from the improvement of material quality with further growth of InGaN active regions in multi-DH LEDs. The integrated PL intensity from the single 6 nm DH LED with 20+20 nm SEI exhibits the trend similar to those for the quad 3 nm DH LEDs with 20+20 nm and 30+30 nm, showing initially superlinear increase at low excitations and it approaches slope one at high excitations.

To experimentally verify the overflow predictions, the integrated electroluminescence (EL) intensity vs. current injection for single 3 nm and 6 nm DH LEDs and quad 3 nm DH LEDs chips with varied SEI thickness was measured (0.1 % duty cycle and 1 kHz to eliminate heating effects). Care was taken to assure invariance in the light collection geometry among all the chips tested. Moreover, the light emission intensity showed less than 5% variation among at least five devices measured on the same wafer.
Figure 3.7: (a) The integrated EL intensity dependence on current density for DH LEDs with varied SEI thickness. (b) The relative EQE vs. injected current density.

Figure 3.7 (a) demonstrates that for the LEDs with 4+4 nm SEI, the EL efficiency of quad 3 nm DH LED is ~3.5 times higher than that of single 3 nm DH LED. It is obvious that the low EQE of the single 3 nm DH LED with 4+4 nm-thick SEI is due to higher
carrier overflow in the thinner active layer facilitating ballistic and/or quasi-ballistic electron transport across the active layer without recombination. This is in good agreement with the calculated electron overflow percentiles shown in Figure 2.3, which predicts substantially higher overflow for the single 3 nm DH LED. Error! Bookmark not defined. The SEI thickness should be larger for thin active region LED like single 3 nm DH to provide hot electrons more time to sufficiently thermalize before being injected into the active region. However, regardless of the active layer design, the total SEI thickness must be kept below the critical thickness above which the active region material quality would degrade noticeably due to strain relaxation. It should also be noted that the agreement between the simulation results of Figure 2.3 and the data presented in Figure 3.7 is satisfactory considering that the calculations assume a constant phonon scattering rate, which would result in underestimation of the overflow percentiles as the phonon lifetime has been reported to decrease with increasing current injection.61

Increase of the SEI thickness from 4+4 nm to 20+20 nm for the single 3 nm DH LED resulted in an enhancement in the peak EL efficiency by nearly 3.5 times, making it comparable to those of the quad 3 nm DH LEDs [Figure 3.7 (a)] and the single 6 nm DH with 4+4 nm SEI. This significant improvement indicates that the electrically injected hot electrons are cooled more efficiently within the thicker SEI. The more visible efficiency roll off in the single 3 nm DH LED both with 20+20 and 30+30 nm-thick SEIs compared to the single 6 nm and the quad 3 nm DH LEDs [Figure 3.7 (b)] suggests larger electron overflow in the thinner active region, which increases at higher injection levels. At the highest injection level employed (550 A/cm², which corresponds to an average carrier density around 5 x 10¹⁸ cm⁻³, estimated using A = 10⁷ s⁻¹ and B = 10⁻¹⁰ cm⁻³ s⁻¹), the
efficiency for the single 3 nm DH LED with 20+20 nm SEI or 30+30 nm SEI is nearly twice that for the single DH LED with 4+4 nm SEI and 55% to 70% of those for single 6 nm and quad 3 nm DH LEDs. It should be noted that thicker, multi-DH or MQW active regions would act as electron coolers naturally. Accordingly, the single 6 nm DH and the quad 3 nm DH LEDs with 20+20 nm SEI show only a moderate increase of 15-20% in the peak EQE compared to those for the LEDs with 4+4 nm SEIs. At injection current densities below 200 A/cm² (the range of the practical interest for LED applications), the quad 3 nm DH and the single 6 nm DH with 20+20 nm SEI show higher relative EQE values due to the reduced overflow as compared to their counterparts with 4+4 nm SEIs and retain about 87% and 79% of their peak EQEs at 150 and 200 A/cm², respectively.

In conclusion, by optimizing the SEI structures, mainly increasing the thickness, the EL efficiency has been improved considerably for single 3 nm DH with 20+20 nm or 30+30 nm SEI owing to the reduced electron overflow. However, further increasing the SEI thickness degraded the material quality which offers lower EL efficiency and PL intensity as well. Further optimization for this structure including slight reducing the total thickness needs to be done.

### 3.4 Delta p-doped barrier in LED active regions

#### 3.4.1 Motivation

Among the mechanisms responsible for the efficiency degradation at high injections, one obvious cause of the efficiency roll-over is the asymmetrical doping in GaN, resulting in an insufficient supply of holes to the active region compared to electrons concentration under electrical injection. Because hole injection lags behind electron injection, the radiative recombination cannot keep up with increasing carrier injection.
These results in either electron escape to $p$-GaN without recombination or electron accumulation in the active region, which reduces radiative recombination efficiency. To address this problem, the efforts were focused on either increasing hole concentration in $p$-GaN or doping barriers in the multi-active region LED structures. As regarding to the second approach, because active regions close to $n$-GaN are lack of holes, carriers recombine mainly in the DH adjacent to $p$-GaN due to the poor hole transport. However, Mg doping in the active region has a detrimental effect on LED efficiency, since Mg acts as luminance “killer” that results in low quantum efficiency of LEDs with doped active regions. So far, there is no experimental report incorporating Mg into the active region without sacrificing the LED efficiency.

Hence, we proposed the employment of $p$-type $\delta$-doping barriers, i.e. Mg was solely doped into barrier during interruption of barrier growth, to improve the hole injection for higher quantum efficiency from one aspect and to eliminate or at least to mitigate possible Mg out-diffusion from barriers to the InGaN active region from another aspect. The experimental data are augmented with numerical simulation of electron and hole concentration in the active regions of LED structures.

### 3.4.2 Experimental procedures

The $c$-plane InGaN LED structures emitting at ~430 nm were grown on ~4 $\mu$m-thick $n$-type GaN templates on sapphire in a vertical MOCVD system. The LED structures, dubbed in the following discussion as samples A, B and C, are shown in Figure 3.8 (a), (b) and (c), respectively.
Figure 3.8: The schematics of hex 3 nm DH LED structures for the improvement of hole injections. Two 6 nm InGaN quantum barriers are located at the n-side for possible p-doping, while the other barriers are 3 nm. (a) In LED A, all the barriers are kept undoped. (b) In LED B, the first 6 nm barrier closest to n-side is p-doped. (c) In LED C, the two 6 nm barriers close to n-GaN were p-doped.

All three LEDs are featured by hex (6x) 3-nm In₀.₁₅Ga₀.₈₅N DH active regions separated by relatively low energy In₀.₀₆Ga₀.₉₄N barriers to enhance the hole transport across the active regions. The three In₀.₀₆Ga₀.₉₄N barriers near p-GaN are 3 nm-thick and the two In₀.₀₆Ga₀.₉₄N barriers near n-GaN are designed to be 6 nm-thick to prevent possible Mg out-diffusion from p-doped barriers into the active regions. The δ-doping with Mg was implemented as follows: after the first 3 nm growth of the 6 nm-thick barrier, the growth was interrupted, and the Cp₂Mg source flew into the growth chamber for 20 s, followed by the growth of remaining 3 nm of the 6 nm barrier. In the reference structure (sample A), all the barriers are kept undoped. In LED B, only the 6 nm barrier closest to n-side is
$p$-doped, while others parts are undoped. In sample C, both 6 nm barriers are $p$-doped. The hole concentration in the doped barrier was estimated to be $4 \times 10^{17}$ cm$^{-3}$, based on delta-doped $p$-GaN deposited in separate experiments. A 15+15 nm-thick SEI consisting of In$_{0.04}$GaN and In$_{0.08}$GaN layers is situated beneath the active region on the n-side to efficiently cool injected hot electrons.~\cite{69,70} A 60 nm Si-doped ($2 \times 10^{18}$ cm$^{-3}$) In$_{0.01}$Ga$_{0.99}$N underlying layer was inserted between SEI and $n$-GaN for quality improvement. The LED structures were completed with 100 nm-thick Mg-doped $p$-GaN layers with $6 \times 10^{17}$ cm$^{-3}$ hole density, as determined from Hall measurements performed on separate samples. Square mesa patterns (400×400 μm$^2$) were formed by conventional photolithography and chlorine based Inductively Coupled Plasma (ICP) etching. Ti/Al/Ni/Au (30/100/40/50 nm) metallization annealed at 860 °C for 60 seconds was used for $n$-type Ohmic contacts, and 5-nm/5-nm Ni/Au film served as the semi-transparent $p$-contacts with 40/50-nm Ni/Au contact pads completed the LED fabrication.

### 3.4.3 Numerical Simulation of delta $p$-doped barrier LEDs

In order to verify the efficiency of delta Mg-doped barriers in improvement of hole injections for LEDs, electron and hole distributions were simulated for the active regions of the investigated LED structures under an injected current density of 100 A/cm$^2$ using a SILVACO ATLAS commercial simulator with simulation parameters appropriate for nitride materials ($A = 10^7$ s$^{-1}$, $B = 10^{-10}$ cm$^{-3}$ s$^{-1}$, and 40% of the theoretical polarization charge was assumed) in Ref. 32.
Figure 3.9: (a) Electron concentration, (b) hole concentration, and (c) band structure) simulated for sample A (black), sample B (red) and sample C (blue) under current injection 100 A/cm² with SILVACO ATLAS with parameters appropriate for nitride materials.

Figure 3.9 (a) exhibits distribution of electrons in the active regions of the three LED structures under investigation for an injection current density of 100 A/cm². As evidenced from the figure, due to efficient cooling of injected electrons in the SEI, the electron
distribution is relatively uniform in sample A. Electron concentrations in the first DH near $n$-GaN are quite similar for all three LEDs, while the second DH in sample B and C exhibits lower electron density compared with sample A. This effect is likely caused by reversed polarization field induced by $p$-type doping of $\text{In}_{0.06}\text{Ga}_{0.94}\text{N}$ barrier, which could also be seen in Figure 3.9 (c). This could also explain the difference in electron concentrations in the third DH, where the electron density is lower in sample C compared to those in sample A and B.

Usually hole transport deep into multilayer active regions of the InGaN-based LEDs is impeded owing to their relatively large effective mass and low mobility. Therefore, without considering hot electron overflow, radiative carrier recombination is limited by the concentration of holes since holes are the minority carriers in the active region under bias. Under these circumstances, as shown in Figure 3.9 (b) for the reference sample A, the hole distribution across the active region is very non-uniform, with the highest concentration in the DH closest to $p$-GaN and a rapid decrease in hole density towards the $n$-side of the LED structure. Therefore, for the conventional LEDs, electron and hole recombination occurs mainly in the DH adjacent to $p$-GaN, while the rest of DHs are lack of holes for recombination. When the InGaN barrier closest to the $n$-GaN is $\delta$-doped with Mg (sample B), the hole concentration in the nearby DH active regions (the first and the second DH) increases significantly, greatly reducing the asymmetry of electron and hole injection and thus enhancing the recombination efficacy. When two 6 nm-thick InGaN barriers are $\delta$-doped (sample C), the hole concentration in DHs near $n$-GaN was further increased, particularly in the DH second closest to the $n$-GaN side, which would further improve the LED output power.
**Figure 3.9** (c) shows the simulation results of the band structure for the three LED structure in the active region under current density 100 A/cm². The δ-doping of the In₀.₀₆GaN barrier closest n-GaN increases the effective height of the barrier in the conduction band (barriers height to quasi-Fermi level is 154 meV, 190 meV, and 192 meV in sample A, B and C, respectively). As a result, the confinement of electrons in the first DH near the n-GaN is improved with δ-doping of the In₀.₀₆GaN barrier.

### 3.4.4 Results and discussion

To demonstrate the effect of p-type delta-doping of barriers on LED performance, integrated EL intensities and relative external quantum efficiency (EQE) were measured for sample A, B, and C under pulsed excitation with 0.1 % duty cycle to eliminate heating effects. As shown in **Figure 3.10** (a), sample B and C exhibit higher integrated EL intensities for high current injection levels (> 50 A/cm²), indicating no obvious material degradation associated with Mg incorporation into the active regions compared to the reference structure. As shown in **Figure 3.10** (b), the peak value of relative EQE for sample B was ~20% higher than that for sample A. The δ-doping of two barriers in sample does not increase the EQE peak value further compared with sample B, which possibly indicates cancelling the positive effect from further improvement of hole injection by reduced active region quality due to the presence of higher amount of Mg in the active region. As an additional benefit of δ-doping, samples B and C suffer less efficiency reduction at high injection compared to sample A (23% for sample A vs. 14% for sample B and C at 400 A/cm²). This finding implies that the improved hole injection partly suppress the efficiency droop at high injection level. EQE peak roll-off in LEDs
with delta-doped barriers shifts towards higher current (80 A/cm$^2$ for sample A, 120 A/cm$^2$ for sample B and C).

![Graph](image)

**Figure 3.10:** (a) The integrated EL intensities and (b) the relative EQE vs. injected current density for sample A, B, and C.

In conclusion, we have shown that δ-doping with Mg of barriers significantly improves the quantum efficiency of LEDs with active regions composed of hex 3-nm DHs. With careful design of the active regions, the improvement in hole injection significantly out-weights the possible degradation of the material quality due to the presence of Mg. With Mg δ-doping of the first barrier on the LED n-side, the relative peak EQE improves by 20% in compared with the reference structure. Doping of the first and second barriers on the n-side does not result in further improvement of relative EQE compared to the case of one doped barrier. This possibly indicates that further improvement in hole injection is canceled by the reduced active region quality due to higher Mg-doping. EQE peak roll-off in LEDs with δ-doped barriers shifts to higher current indicating the reduction of
asymmetry in injection of electrons and holes with the employment of δ-doping. The both LED structures with Mg delta-doped barriers effectively suppress the efficiency droop at high injection level compared to the counterpart with un-doped barriers.

3.5 Investigation of GEI LEDs

We examined LED structures with different active-region and SEI designs and aimed at reducing the electron overflow (better efficiency retention at high injection current density) as well as improving the quantum efficiencies. In Chapter 2, we already theoretically investigated GEI and SEI on the effects of electron cooling, and GEI exhibits better performance in suppression of electron overflow than SEI with the same total electron injector thickness. In this realm, 6x2 nm and 6x1.5 nm MQWs with GEI: 20 nm were analyzed and compared with the best LEDs with two-step SEI designs.

![Graph showing IQE vs Excitation density for different SEI designs](image)
We can see that, in Figure 3.11 (b), with the same active region thickness, hexa 2 nm MQW LED shows 20% higher EL efficiency than quad 3 nm DH LED even MQW LED was employed with thinner electron injector (20 nm versus 40 nm). If compared with single 6 nm with 20+20 nm SEI, hexa 2 nm MQW shows 10% higher peak EQE value and much less efficiency droop at high injection up to 300Acm$^{-2}$. Moreover, hexa 1.5 nm MQW LED shows nearly double EQE peak value compared with that of quad 3nm DH LED. The mechanism of this great EL intensity improvement is still under investigation.
3.6 Injection-dependent radiative recombination coefficient (B) of single and multi active layer DH LEDs

The radiative recombination coefficient, B, is dependent on injection through a variety of processes. Change in junction temperature due to Joule losses is one such process as B coefficient is inherently temperature dependent. Carrier lifetime, which is related to the B coefficient, increases with temperature which implies that the B coefficient reduces with increasing junction temperature exacerbated by limitations in heat transfer. The B coefficient is also dependent on injection due to the skewed band edges in the active region of LEDs and also on the total number of states that are available. In biaxially strained InGaN/GaN layers, the mismatch strain induces a polarization field (allowed by the wurtzite-structure lattice symmetry) along the growth direction. This strain induced piezoelectric field and spontaneous polarization field tilts the potential profile, resulting in triangular quantum wells in the hetero-interfaces of active regions. Consequently, the optical transition energy is reduced. Therefore, with increasing current injection, a blueshift will occur and the magnitude of the blueshift depends on the active layer thickness. The simulated potential profiles, using the commercial Silvaco ATLAS software package, are shown in Figure 3.12 (a) and (b) for single 3 nm and the 9 nm DH LEDs, respectively, at different injection current densities. Note that a factor of 0.4 was used for the polarization induced charge for both electrons and holes at the interfaces in generating Figure 3.12 (a) and (b). In a thick active layer like single 9 nm DH, electron and hole wavefunctions are widely separated at low injection levels, which results in reduced radiative recombination rate and relatively low EQE. We have to note that the available states of the triangular potential well are not completely filled at low current
injection. With increasing the injection level, the triangular region will be fully occupied due to relatively low density of states. With further increase in injection, the quasi-continuum states followed by the three dimensional states in the DH active region will begin to fill, leading to higher recombination rate and thus higher relative EQE.
Figure 3.12: Energy band edge profiles simulated for (a) 3 nm DH and (b) 9 nm DH LEDs with 4×4 nm SEIs at different injection current densities. Peak emission energy shift as a function of injection current density from (c) EL measurements and (d) Silvaco simulations for DH LED structures with various active regions.

Figure 3.12 (c) shows the observed peak emission energy shift as a function of injection current density from electroluminescence (EL) measurements. Upon current injection, the polarization fields in the strained InGaN DH layer are screened by free carriers, which weakens the quantum confined Stark effect (QCSE), and then increases the transition energy resulting in the blueshift in emission. Larger peak energy shifts were observed with injection when the DH thickness is increased from 3 nm to 9 nm, as presented in Figure 3.12 (c). The entire DH LEDs exhibit a large portion of the energy shift at low injection levels (< 50 A/cm²) followed by a slower shift with further increase in injection. This is attributed to the presence of triangular potential wells at the interfaces, which are filled rather rapidly even at relatively low injection levels causing the large initial blueshift. Recombination from the higher lying quasi-continuum states and 3D states in
the DH active region start to contribute to emission with increased injection, resulting in the slower blueshift observed.

For single 6 nm, dual 6 nm, and single 9 nm DH LEDs, a significant 25 meV energy shift was observed with initial increase of current density to 40 A/cm², while only a 8 meV shift is evident for the 3 nm DH LED within the same current density range. Figure 3.12 (d) shows the simulated peak energy shift as a function of current density obtained using Silvaco ATLAS. The polarization charge densities of 7.7x10^{12} cm⁻² and 3.8x10^{12} cm⁻² have been used for the interfaces between the In₀.₀₁Ga₀.₉₉N barrier and the In₀.₁₅Ga₀.₈₅N active layer, and the In₀.₁₅Ga₀.₈₅N active layer and the In₀.₀₈Ga₀.₉₂N SEI, respectively, which are within the range of reported values ⁷⁴, and provide energy shifts in the range comparable to those observed experimentally. The tendency is consistent with the experimental results except for the single 9 nm DH and 11 nm DH structure. This observed discrepancy is likely due to the strain-driven material quality degradation for thick single DH active region (9 nm and 11 nm DH) neglected in simulations. We should also note that varying the polarization charge in the simulations changes the absolute energy shift, but does not affect the peak energy shift dependence on the active layer thickness.

We now discuss the electron and hole wavefunction overlap in the context of radiative recombination $B_{eff}$ coefficient as a function of injection current density in a system with confinement along the z-direction (growth direction). Following the formalism of Ref. 75, in the realm of Fermi’s Golden Rule, the spontaneous transition rate from a group of initial states i in the conduction band to a group of final states f in the valence band separated by a transition energy $\hbar \omega$ can be expressed as
\[
T_{\rightarrow f} = \frac{2\pi}{\hbar} \left( \frac{eA_0}{2m_0} \right)^2 |M|^2 \left| \int \psi^*_f(z) \psi_i(z) dz \right|^2 \rho_r(\hbar \omega) F(\hbar \omega)
\]

(3-1)

where \( A_0 \) is the magnitude of the sinusoidal local vector potential, \( e \) is the electron charge, \( m_0 \) is the free electron mass, \( M \) is the in-plane momentum matrix element, \( \rho_r(\omega) \) is the reduced density of states, \( \omega \) is the transition energy, \( F = f_c(1-f_v) \) is the Fermi factor given in terms of the Fermi functions for the conduction \( (f_c) \) and valence bands \( (f_v) \), \( \Psi(r) = \psi(z) \phi(r_{xy}) \) are the envelope functions representing the wavefunctions. As noted in Eq 3-1, the spatial overlap between the electron and hole wavefunctions \( (\psi_e \text{ and } \psi_h) \) is obviously imperative, and the radiative recombination rate is proportional to the squared overlap integral when electrons and holes are confined in the \( z \)-direction.

For quantum-confined structures, it has been suggested that low-dimensional equivalents of the bimolecular radiative recombination B coefficient should be introduced to eliminate the artificial dependence of the radiative recombination current on size, such as the active region width in two-dimensional (2D) systems.\(^7\) By defining the spontaneous transition rate as \( T_{\text{spont}} = B_{2D} n_{2D} p_{2D} \) where \( n_{2D} \) and \( p_{2D} \) are the 2D electron and hole densities, respectively, the 2D \( B \) coefficient in InGaN quantum wells with confinement along the \( z \)-direction can be expressed in terms of the momentum matrix element of Eq 3-2:
where $n$ is the refractive index, $\varepsilon_0$ is the permittivity of free space, $c$ is the speed of light, $\hbar \omega$ is photon energy, $k_B T$ is the thermal energy, and $m_e^*$ and $m_h^*$ are the electron and hole effective masses. The momentum matrix element $M$ can be obtained from the in-plane interband transition matrix element (for polarization within the plane), $P_{cv} = 2|M|$, which has been determined from the absorption measurements for binary constituents InN and GaN.\textsuperscript{77} Using $P_{cv} = 9.6 \times 10^{-20}$ g cm/s obtained from linear interpolation for the required composition, we calculated the $B_{2D}$ coefficient to be $1.8 \times 10^{-4}$ cm$^2$s$^{-1}$ for an In$_{0.15}$Ga$_{0.85}$N active region assuming full overlap of electron and hole wavefunctions. For 3D rate equation, applicable to 3D DH structures, the 2D $B$ coefficient should be multiplied by the active region thickness, $L_z$. To test this approach and the validity of the 2D approximation, the 3D limit for the $B$ coefficient for In$_{0.15}$Ga$_{0.85}$N was also calculated from the following equation\textsuperscript{78}

$$B_{3D} = \frac{e^2 n}{m_e^2 c^3 \hbar^2} \left( \frac{2 \pi \hbar^2}{k_B T} \right)^{3/2} \times \rho_e (\hbar \omega),$$

(3-3)

where $\vec{m}_{x,y,z} = m_{e(x,y,z)} + m_{h(x,y,z)}$. The value of 3D $B$ coefficient derived from Eq. 3-3 is $5 \times 10^{-11}$ cm$^3$s$^{-1}$ for In$_{0.15}$Ga$_{0.85}$N. This value is smaller than that obtained using $B_{2D} L_z$ even for the thinnest active region with $L_z = 3$ nm investigated in this study. We, therefore, assume that all LEDs with active region widths of 3 nm and above exhibit 3D behavior but with an electric field along the growth direction reducing the spatial
overlap of charge carrier distributions in the active region. Therefore, the injection dependent overlap integral of the electron and hole wavefunctions should be incorporated into the calculation of the 3D $B_{\text{eff}}$ coefficients using $5 \times 10^{-11} \text{ cm}^3/\text{s}$ as the upper limit for full overlap:

$$B_{\text{eff}} = \left(5 \times 10^{-11}\right) \int_0^h |\psi_h(z)|^2 |\psi_e(z)|^2 \, dz$$

(3-4)

Figure 3.13: Calculated coefficients $B_{\text{eff}}$ of (a) single DH, and (b) multi DH LEDs, calculated using squared overlap integrals of electron and hole wavefunctions (proportional to radiative recombination rate) within the active region as a function of current density using SILVACO ATLAS software package. The SEI layer thicknesses are provided in the legends in nm units.
Figure 3.13 represents the simulated bimolecular recombination coefficients, $B_{\text{eff}}$, derived from the transition matrix element and the simulated squared overlap integrals of the electron and hole wavefunctions in the single and multi DH active regions. It is apparent from Figure 3.13 that the radiative recombination coefficient, instead of being constant as assumed,\textsuperscript{79} in fact depends on injection and particulars of the active-region.\textsuperscript{80} The $B_{\text{eff}}$ coefficient initially increases with injection and then tends to saturate at high injection levels as the flat band condition is approached. Thinner active layers (single 3 nm DH) exhibit larger $B_{\text{eff}}$ values at low injections, which is attributed to their relatively larger spatial overlap of the electron and hole wavefunctions. At \( \sim 200 \text{ A/cm}^2 \), the single 3 nm DH LEDs exhibit about 37\% and 60\% higher squared overlap integral value compared to the single 6 nm and the single 9 nm DH LEDs, respectively. The change in the $B_{\text{eff}}$ coefficient with injection is very small for the single 3 nm DH LEDs, while that for wider DHs exhibits substantial increase of $B_{\text{eff}}$ coefficient within injection in the range of 0 A/cm\(^2\) to 500 A/cm\(^2\). The smaller $B_{\text{eff}}$ coefficients in wider active regions are, naturally, attributable to increased spatial separation of electrons and holes caused by the polarization fields.\textsuperscript{81}

It is worth noting that wider SEIs give rise to larger overlap integral for otherwise the same active layer thickness. The difference in the overlap integrals for the single and the dual 6 nm DH LEDs with 4+4 nm and 20+20 nm SEIs exhibiting 18\% and 22\% larger $B_{\text{eff}}$ coefficients, respectively, at 100 A/cm\(^2\). When injection approaches 500 A/cm\(^2\), this difference is somewhat reduced to 13\% and 20\% for the LEDs with 4+4 nm and 20+20 nm SEIs.
It has been reported that radiative and nonradiative coefficient is proportional to square of electron and hole wave functions overlap.\(^{82}\) In order to get the dependence of radiative and nonradiative coefficient on temperature, we can use the radiative and nonradiative decay time to extract them. Our assumption is that radiative process is dominant at low temperatures, \(i.e.\) the internal quantum efficiency is 100\% at low temperature. The PL intensity is proportional to the ratio of nonradiative (\(\tau_{nr}\)) decay times to total decay times as following:

\[
I(T) \propto \frac{\tau_{nr}}{\tau_r + \tau_{nr}} \propto \frac{1}{1 + \frac{\tau_r}{\tau_{nr}}} \\
\tag{3-5}
\]

where \(I(T)\) is PL intensity as a function of temperature. If we normalize the PL intensity to unity at 10 K, then we can obtain the following equation:

\[
I(T) = I(10\,K) \times \frac{1}{1 + \frac{\tau_r}{\tau_{nr}}} \\
\tag{3-6}
\]

where \(I(10\,K)\) is the PL intensity at 10 K. This quenching behavior which is typical for nitride semiconductors is caused by thermal activation of nonradiative recombination channels and/or localization energy above a certain temperature.

Note that the measured PL decay time can also be expressed in terms of radiative (\(\tau_r\)) and nonradiative decay times (\(\tau_{nr}\)) as
\[
\frac{1}{\tau_{PL}} = \frac{1}{\tau_r} + \frac{1}{\tau_{nr}}
\]

(3-7)

where \(\tau_{PL}(T)\) is the total or effective PL decay time and also the quantity obtained from fitting the PL transients.

Using the Eq. 3-6 and 3-7, one can reach the temperature dependence of the radiative and nonradiative decay times as a function of temperature.

Figure 3.14: The PL decay time (\(\tau_{PL}\)), radiative decay (\(\tau_r\)) time and nonradiative decay time (\(\tau_{NR}\)) for the hexa 1.5 nm (a), 2 nm (b) and 3 nm (c) LEDs as a function of temperature

The results are shown in Figure 3.14. As can be seen, radiative recombination is dominant process at low temperature. Up to 100 K radiative decay time is almost independent of temperature, indicative that radiative decay time is determined by the contribution of bound excitons. Nonradiative recombination rate is very low below 100 K, because bound excitons will mainly have radiative recombination at low temperature,
while nonradiative recombination centers (deep level defects in the layer) have negligible effect at low temperature. Only a decrease of the emission intensity can be expected with increasing temperature because of ionization of the bound excitons at this temperature range. The carriers are thermally activated and reached defects where they can recombine nonradiatively above 100 K for all samples. The radiative to nonradiative transition point at which the nonradiative recombination rate starts to exceed radiative recombination rate, appeared at almost same temperature for all LEDs. With increasing temperature, bound excitons break out and free electrons and holes recombination dominant in carriers recombination. Therefore, nonradiative lifetime decreases with increasing temperature. The reason for the increase in radiative lifetimes with increasing temperature could be attributed to the decreasing probability of overlapping of electron and hole wave function because of the screening of the phonon emission.

Using the Eq. 3-8 and 3-9, we can reach the temperature dependence of the nonradiative and radiative coefficient.

\[ A = \frac{1}{\tau_{nr}} \]

(3-8)

\[ B = \frac{1}{\tau_{r,n}} \]

(3-9)

It is clear that A coefficient increases with increasing temperature and nonradiative recombination is dominant in room temperature. In contrast, B coefficient decreases with increasing temperature and radiative recombination is dominant in low temperature.
3.7 Investigation of layer quality on semi-polar GaN

3.7.1 Motivation
As already mentioned earlier, in biaxially strained InGaN/GaN layers, the mismatch strain induces a polarization field (allowed by the wurtzite-structure lattice symmetry) along the growth direction. This strain induced piezoelectric field and spontaneous polarization field tilts the potential profile, resulting in triangular quantum wells in the hetero-interfaces of active regions. Especially in a thick active layer, electron and hole wavefunctions are widely separated at low injection levels, which results in reduced radiative recombination rate and relatively low EQE. Therefore, nonpolar and semipolar GaN substrates, where the spontaneous polarization field is zero or much weaker than polar c-GaN, have prospective use in optoelectronic devices due to larger wavefunction overlap of electrons and the holes in active regions increasing the quantum efficiency.

3.7.2 Experimental procedure
To achieve (1 1 0 1) GaN film surface parallel to the substrate, Si (001) substrates that are miscut 7° toward the Si ⟨1 1 0⟩ direction were used. The substrates were patterned to form grooves aligned parallel to the Si ⟨1 1 0⟩ direction as described elsewhere.83,84 The top terraces were 3 μm wide, while grooves were either 10 or 3 μm wide (hereafter, 3 μm x 10 μm and 3 μm x 3 μm patterns, respectively). A 50-nm-thick AlN layer was grown by MOCVD to serve as a seed layer for GaN growth. SiO₂ mask layer was formed on one of the sidewalls of the groove, and subsequent growth of GaN started on the opposite side of the groove. The substrates were then reloaded into the MOCVD chamber and GaN
growth was performed with the use of trimethylgallium (TMG) and NH$_3$ as sources of Ga and N, respectively. Semi-polar GaN layers were grown at a chamber pressure of 200 Torr and high NH$_3$ flow rates (>2000 sccm), which were found to be favorable for the (1101) facet formation. The growth initiated on Si {111} sidewalls free from SiO$_2$ and then advanced laterally first along the GaN [0001] $c^+$ direction and then additionally along the [0001] $c^-$ direction after the vertical growth advanced above the Si (001) terraces, which are shown in Figure 3.14.

![Figure 3.15: (a) Cross-sectional SEM image of coalesced semi-polar GaN layers with 3 um x 3 um pattern; (b) inclined SEM image of non-coalesced semi-polar GaN layers with 3 um x 10 um pattern.]

**3.7.3 Results and discussion**

A series of optical measurements were performed to investigate the quality of semipolar (1101) GaN grown on patterned Si (001) substrates. A $c$-plane (0001) GaN/sapphire and a $c$-GaN layer grown on the state-of-art GaN template using in-situ epitaxial lateral overgrowth with SiNx nano-network to block dislocation propagation (referred as nano-
ELO layer) were measured as reference samples. Figure 3.15 compares the room temperature PL spectra from the c-plane GaN film on sapphire, nano-ELO layer, non-coalesced and coalesced semipolar (1\(\bar{1}01\)) GaN layers. We can see that the intensity of near-band-edge (NBE) from non-coalesced (1\(\bar{1}01\)) GaN is comparable to that of the c-plane layer grown on sapphire. The PL intensity of coalesced GaN layer is much weaker, which can be due to defects formed in the layer as a result of coalescence and relatively larger total area of the defective c\(^{-}\) wings.\(^{85}\)

Figure 3.16: Steady-state room-temperature PL spectra of c-plane GaN films grown on sapphire and (1 100) m-plane and (1101)-oriented GaN layers grown on the Si patterned substrates.

Normalized TRPL data were fit using a biexponential decay function \(A_1e^{-t/\tau_1} + A_2e^{-t/\tau_2}\), where \(A_1\) and \(A_2\) are the amplitudes of the slow and fast decay components with representative time constant \(\tau_1\) and \(\tau_2\), respectively. Figure 3.16
compares room-temperature excitation density dependent TRPL for c-plane nano-ELO GaN on sapphire and non-coalesced (1 ̅101)GaN on Si (001), and Table 3-3 lists the fitting parameters for c-plane and (1 ̅101)GaN layers. The normalized TRPL data were fit by using the bi-exponential decay function. As seen from Figure 3.16 and Table 3-3, the slow decay component τ₂ for both layers becomes longer with increasing excitation power density. It indicates that nonradiative and radiative recombinations compete at low excitations, while radiative recombination becomes dominant at high excitation density. The decrease of radiative recombination time with increasing excitation power density is attributed to increased electron and hole overlap resulting in faster radiative recombination. The slow decay component for the non-coalesced layer is as long as 1.98 ns, which is even longer than that of the state-of-the-art nano-ELO GaN layer (1.15 ns).

This long slow decay constant is indicative of high optical quality of the noncoalesced layer and lower density of extended and point defects compared to coalesced layer. In the case of semipolar GaN, regions of high quality c⁺ wing regions are probably responsible for the long slow decay components τ₂ at high excitation levels.

Figure 3.17: Excitation density dependent room-temperature TRPL for (a) polar
(0001) nano-ELO GaN film on sapphire and (b) semipolar non-coalesced semi-polar GaN layer on Si.

Table 3-3: PL decay times and amplitude ratios obtained from biexponential fits.

<table>
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<tr>
<th>Excitation (kW/cm²)</th>
<th>τ₁ (ns)</th>
<th>τ₂ (ns)</th>
<th>A₁/A₂</th>
<th>τ₁ (ns)</th>
<th>τ₂ (ns)</th>
<th>A₁/A₂</th>
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<td>0</td>
<td>0.39</td>
<td>1.90</td>
<td>1.17</td>
</tr>
<tr>
<td>0.32</td>
<td>-</td>
<td>1.15</td>
<td>0</td>
<td>0.40</td>
<td>1.98</td>
<td>1.19</td>
</tr>
<tr>
<td>(0001) non-coalesced layer</td>
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80
Chapter 4  
Quantum efficiency enhancement in GaN-based VCSELs

4.1 Hybrid VCSELs

4.1.1 Brief introduction to hybrid VCSELs

Similar with widely used GaAs-based VCSELs, GaN-based VCSELs are expected to show various advantages over the edge-emitting lasers. Unfortunately, several obstacles make the progress of GaN-based VCSELs relatively slow. The key problems limiting the development of VCSELs are the difficulty in growing high quality and high reflectivity distributed Bragg reflectors (DBRs) and to obtain high quality cavity active region which is embedded within two highly reflectivity DBRs. High quality and high reflectivity DBRs have a critical role in order to achieve threshold condition in VCSELs due to the relatively short gain region.

In general, there are three types of III-Nitride DBRs, including AlN/GaN, Al\textsubscript{x}Ga\textsubscript{1-x}N/Al\textsubscript{y}Ga\textsubscript{1-y}N, and Al\textsubscript{x}In\textsubscript{1-x}N/GaN DBRs. The AlN/GaN DBRs have the highest refractive index contrast (2.185 for AlN and 2.512 for GaN at wavelength 430 nm) among all the three kinds of DBRs and provide high reflectivity together with a relatively large stop-band width. However, the large lattice mismatch between AlN and GaN results in accumulation of strain during growth and formation of cracks. These cracks seriously deteriorate the reflectivity of the DBRs in addition to the quality of the following active region. Unlike the AlN/GaN DBRs, which suffer from cracking, the strain can be reduced in Al\textsubscript{x}Ga\textsubscript{1-x}N/Al\textsubscript{y}Ga\textsubscript{1-y}N DBRs with appropriate adjustment of Al/Ga composition. However, with Al\textsubscript{x}Ga\textsubscript{1-x}N/Al\textsubscript{y}Ga\textsubscript{1-y}N DBRs, the refractive index contrast decreases between the pairs of layers which results in a reduced
stop-band width and thus the necessity for increased number of pairs for high reflectivity. In contrast, lattice matched $\text{Al}_{0.83}\text{In}_{0.17}\text{N}/\text{GaN}$ DBRs, which could avoid subsequent layer degradation, were reported with cavity quality factors up to 6400.\textsuperscript{88} However, relatively lower refractive index contrast (7-8%) in $\text{Al}_{0.83}\text{In}_{0.17}\text{N}/\text{GaN}$ DBRs would result in narrower stop-band width and thereby increased number of pairs for high reflectivity.\textsuperscript{89} Besides the three kinds of III-Nitride DBRs, another possible solution for VCSELs is to use vertical cavity with bottom dielectric DBRs instead of bottom AIN/GaN DBRs. However, the typical employment of bottom dielectric DBRs of VCSEL requires the removal of majority of GaN substrate with a precise control of remaining cavity length,\textsuperscript{90} which is not practical for wide application. In addition, the VCSELs performance also depends strongly on the active region design and the promising InGaN active region design, optimized from LEDs experiments, would be employed in VCSEL structures to check the cavity performance. Therefore, the investigation of high quality and high reflectivity DBRs growth and followed high quality active region growth is imperative for high performance VCSEL devices.

### 4.1.2 Experimental procedures

In order to obtain crack-free AIN/GaN DBRs favorable for high reflectivity and wide stop-band for GaN-based VCSELs, MOCVD growth for AIN/GaN DBRs on GaN/c-sapphire template was investigated. For the purpose of releasing strain accumulation in layers growth, 5.5 pairs AIN/GaN super-lattice layers were inserted in every 5.5 periods of GaN/AIN DBRs deposition. The whole growth in MOCVD chamber was kept at low pressure (76 Torr) in N$_2$ environment.\textsuperscript{91} The reflectivity of obtained crack-free 29 pairs AIN/GaN DBRs and cross-sectional image of AIN/GaN DBRs from JEOL scanning
electron microscope (SEM) is shown in Figure 4.1. Because the DBRs is formed of AlN and GaN layers having large refractive index difference, the reflectivity above 97% with around 18 nm stop band with central wavelength at 435 nm was achieved with 29 pairs of AlN/GaN DBRs.

![Graph and Microscope Image]

Figure 4.1: (a) Reflectivity of 29 pairs crack-free AlN/GaN DBRs on GaN template, (b) cross-sectional images of 29 pairs crack-free AlN/GaN DBRs from scanning electron microscope.

The VCSEL structure, shown in Figure 4.2, contains the two active regions embedded between the top and bottom DBRs, with each of the active regions consist of hexa 3 nm
In\textsubscript{0.15}Ga\textsubscript{0.85}N DHs separated by 3 nm In\textsubscript{0.01}Ga\textsubscript{0.99}N barriers. The two active regions were separated by 132 nm (\(\lambda\)) In\textsubscript{0.01}Ga\textsubscript{0.99}N underlying layer. The designed cavity length is 2.5 \(\lambda\) at 410 nm for the VCSEL structure. The VCSEL structure was completed with 13 pairs SiO\textsubscript{2}/SiN\textsubscript{x} dielectric DBRs as the top DBRs, deposited at 300 °C using ultra high vacuum chemical vapor deposition (UHVCVD). High reflectivity (> 99.5%) with ~80 nm stop-band width was achieved with a designed stop-band central wavelength of 430 nm for top dielectric DBRs.

![Diagram of VCSEL structure](image)

**Figure 4.2**: The structure schematic of the vertical cavity surface emitting laser: 2.5\(\lambda\) cavity with two hexa 3 nm In\textsubscript{0.15}Ga\textsubscript{0.85}N DHs separated by 132 nm (\(\lambda\)) In\textsubscript{0.01}Ga\textsubscript{0.99}N underlying layer grown on bottom 29 pairs crack-free AlN/GaN DBRs on 4 µm GaN template and completed with 13 pair top SiO\textsubscript{2}/SiN\textsubscript{x} DBRs.

### 4.1.3 Results and discussions

**Figure 4.3** shows the reflectivity spectrum for the full vertical cavity (black), reflectivity spectrum for the bottom AlN/GaN DBRs (red) and PL spectrum (blue) emitting between top dielectric and bottom semiconductor DBRs. The micro-PL was measured using a
frequency-doubled Ti:Sapphire laser (380 nm wavelength). The inset in Figure 4.3 shows the micro-PL for the cavity mode having Q-factor of 1300. However, not all the regions on this sample showed this high Q-factor, which results in part from the non-uniform active region quality on the wafer. During AlN/GaN bottom DBRs growth, owing to its large lattice mismatch, a large density of dislocations was generated on the wafer which greatly degrades the active region quality. In order to achieve higher Q-factors, further optimization efforts focused on achieving high reflectivity bottom DBRs, improving the active region quality, and achieving sharper cavity interfaces were necessary.

![Reflectivity spectrum](image)

Figure 4.3: The reflectivity spectrum (black) for the full vertical cavity, reflectivity spectrum for the bottom AlN/GaN DBR (red) and photoluminescence spectrum (blue).

As performed earlier for the semipolar InGaN active region grown on noncoalesced GaN layer on patterned Si, in order to gain insight into the distribution of active region emission from the full cavity, room temperature NSOM was performed (Figures 4.4).
Figures 4.4 (a) represents the AFM image of the full cavity (13 pairs top dielectric DBR surface) for the scan area 50x50 µm² and Figures 4.4 (b) represents the corresponding active region emission for the scanned area. The PL emission from the microcavity is found to be not uniform over the scanned region. The reason might be different structural quality or differences in the cavity length in this region.

Figure 4.4: Room temperature NSOM results for the microcavity structure with bottom semiconductor and top dielectric DBRs (a) height image (b) PL intensity mapping.
4.2 GaN-based VCSELs with both dielectric DBRs

4.2.1 Motivation

The InGaN/GaN laser structure with bottom AlN/GaN DBRs suffers from the degradation of active region quality. Dislocations generated during the AlN/GaN DBRs growth would greatly degrade layer quality of the active region and result in current leakage under electrical injection, which would severely affect laser performance. One possible solution is to use vertical cavity with both dielectric DBRs instead of hybrid cavity with bottom AlN/GaN DBRs. However, the typical employment of dielectric DBRs in both sides of VCSEL cavity requires the removal of majority of GaN substrate with a precise control of remaining cavity length. Therefore, in order to obtain high quality of active region with high reflectivity dielectric DBRs, epitaxial lateral overgrowth (ELO) of GaN using dielectric DBRs (SiO$_2$/SiN$_x$ masks) as the bottom reflector instead of the AlN/GaN bottom DBRs has been proposed. The “wing region” (overgrown GaN layer) would have much lower density of dislocations compared with “window region”, which would greatly improve the active region quality of the laser cavity. Moreover, due to the larger refractive index difference between SiO$_2$ and SiN$_x$, higher reflectivity (more than 99.5%) and much wider stop band (more than 80 nm) could be achieved easily for the dielectric DBRs compared with the AlN/GaN DBRs, which could benefits the laser cavity design. Therefore, GaN-based VCSELs with both dielectric DBRs are promising for improvement of VCSEL performance.
Figure 4.5: The reflectivity spectrum of bottom 13 pair SiO$_2$/SiN$_x$ bottom DBR.

4.2.2 Experimental procedures

4.2.2.1 Growth procedures of ELO-GaN for VCSEL

For the GaN-based VCSELs with both dielectric DBRs, it the critical to achieve appropriate ELO wings for VCSELs with both dielectrics DBRs. For the purpose of devices fabrication for electrical injection, the window width is required to be much narrower compared with separation width, so that much longer ELO wing size could be achieved. Nevertheless, for the ELO growth with larger fill factor (wing-to-window ratio), the tilt of surface of the ELO-GaN with respect to underlying GaN layer becomes more apparent, which would pose a major obstacle for the VCSEL structure. Therefore, it is desirable to maximize the lateral versus vertical growth ratio and reduce wing tilt of the ELO-GaN. A large lateral growth rate and a horizontal wing with greatly reduced wing tilt were achieved by employing the NH$_3$ flow modulation technique. In this method,
MOCVD chamber growth pressure was kept at 30 Torr to improve the lateral growth ratio of GaN. The NH₃ flow was interrupted for 15 s after every 20 s of regular GaN growth, while trimethylgallium (TMGa) flow rate was kept constant. This growth was performed using a dielectric DBRs mask with 4 μm-wide stripe-shaped windows separated by 32 μm and aligned along the GaN <1100> direction to enhance the lateral growth ratio. The cross-sectional images of the ELO-GaN stripes were measured with SEM, shown in Figure 4.6.

![Figure 4.6: Cross-sectional SEM images of ELO-GaN grown (a) with constant NH₃ flow for 3 hours and (b) with NH₃ flow modulation for 2 hours. Wing tilt angle in case of NH₃ flow modulation was measured to be lower than 0.1° whereas that in the case of constant NH₃ flow was 5°.](image)

From Figure 4.6, it is clear that ELO-GaN grown with NH₃ flow modulation had larger lateral growth rates and much flatter surfaces than those with constant NH₃ flow. Lateral advancement of the ELO wings with NH₃ flow modulation for 2 hours was large enough (13 μm) for the device fabrication, while the ELOs with constant NH₃ flow had the same lateral size after a longer 3 hours growth. Moreover, the modulation method resulted in a much smaller wing tilt (< 0.1°) than the standard one (5°). This improvement is due to enhanced migration of Ga atoms on the (0001) plane of ELO-GaN surface, since the Ga atoms are more freely to move toward wing region with pulsed NH₃ flow profile. For the further improvement of the lateral vs. vertical growth ratio of ELO-GaN, effects of
growth parameters, such as temperature, NH$_3$ flow rate, TMGa flow rate, need be investigated in order to achieve appropriate ELO-GaN for laser diodes.

As reported in the first annual report, in order to get quality improvements of active region, epitaxial lateral overgrowth (ELO) of GaN using dielectric DBR masks as the bottom reflector is worthy for investigation. The “wing region” (overgrown GaN) would have much lower density of dislocations compared with the window region. In terms of efficiency as well as device application, it is critical to maximize the lateral versus vertical growth ratio and achieve a flat surface and wide enough for ELO-GaN wing region. To achieve the goal, NH$_3$ flow modulation technique is employed to enhance the lateral overgrowth, since it could improve the migration of Ga atoms on the (0001) plane of ELO-GaN surface. In this method, interruption of NH$_3$ flow was inserted for a given time (15, 25, 30 s were test) during growth for every period (20 s) of normal GaN growth, while TMGa (trimethylgallium) flow rate was kept constant. This growth was performed using a growth mask with varied width such as 2, 3 and 4 μm-wide stripe-shaped windows separated by 34, 33 and 32 μm and aligned along the GaN $<\bar{1}\bar{1}00>$ direction, respectively. We tried to optimize the growth parameter such as periods of ammonia on/off time and window size for ELO stripe to get the improvement of ELO-GaN growth results. From the cross-section image of scanning electron microscopy (SEM) in Fig. 22, we successfully improve the ELO-GaN lateral/vertical ratio from 1 in Figure 4.7 (a) to around 4 Figure 4.7 (b) after 1.5 h growth.
Figure 4.7: Cross section pictures of ELO growth for (a) ammonia on/off time 20 s/15 s with TMGa: 20 sccm and (b) ammonia on/off time 20 s/25 s with TMGa: 12 sccm

4.2.2.2 Etching procedures for ELO-GaN for VCSEL
After the optimization of the ELO-GaN growth conditions, effectively etching of ELO-GaN layer would be employed to reduce the ELO thickness in order to achieve an appropriate cavity length. Due to the chemical inertness and relatively high bond energies, GaN materials are resistant to conventional wet etching, and available wet etches process is rarely attainable with photolithography. Therefore dry etching techniques with high plasma density and ion energies, such as reactive ion etch (RIE) or inductively coupled plasma (ICP) etching process based on different gas mixtures and conditions, are essentially important and extensively used for the etching of GaN films and GaN-based device structures. Usually for normal GaN film’s etching, high etch rates and anisotropic profiles for making mesa-patterns could be achieved if the etching conditions are precisely controlled. Although this is convenient for mesa-pattern fabrication, the problem of the etching of ELO-GaN has so far not been sufficiently explored in the literatures. Variations in the quality of the window region (seed GaN) and wing region (overgrown GaN) of ELO-GaN pattern present unique challenges for dry etching.
processes of ELO-GaN. Therefore, it is necessary to investigate the etching conditions to reduce ELO-GaN thickness efficiently and achieve smooth ELO-GaN surface. In this section, we investigate the characteristics of the etching process of ELO-GaN by using a Cl₂/Ar/SiCl₄ gas mixture, for potential application in fabricating ELO-GaN based VCSEL and polariton laser structure. The use of the Cl₂ for ICP etching of GaN implies an enhancement of the chemical component to get a high etch rate. The use of Ar is for physical etching component to balance the chemical etching component. SiCl₄ serves as a passivation layer on the etched side walls. The crystal quality of the ELO-GaN was characterized by X-ray diffraction (XRD). The effects of etching conditions were investigated by scanning electronic microscopy (SEM) and Atomic force microscopy (AFM). In this work an optimization of an ICP dry etching method is made to get smooth surface, minimum damage, a relatively high etch rate and appropriate profiles.

Regular 4 μm GaN template with a low-temperature GaN buffer layer was grown MOCVD system on (0001) sapphire. After template growth, SiO₂/SiNx DBRs (center wavelength target at ~410 nm and stop-band ~80 nm with reflectivity >99.9%) were deposited by PECVD on the GaN template as bottom reflector. Fabrications were applied to open the stripe patterns along the [1100] direction of the underlying GaN template. The window widths were 4 μm with a periodicity of 36 μm. ELO-GaN was grown overlying the dielectric mask with pulse NH₃ treatment and constant TMG flow. After growth, the ELO-GaN layers had a flat plane (0001) and didn’t coalescence. The tilt of the c-axis on the mask area was investigated by Philips X’Pert-MPDTM X-ray rocking curves (XRCs). The XRCs of the (0002) reflection of the ELO-GaN layer was measured at ϕ = 90º, where ϕ is the azimuth angle between the stripe direction of the mask and the scattering plane.
Then the ELO-GaN sample was dry etched by ICP. The etchant gases were a Cl\textsubscript{2}/Ar/SiCl\textsubscript{4} gas mixture. All the gases were introduced into the reactor chamber through independent mass flow controllers (MFCs) that can control the flow rate of each gas. The flow rates of Ar and SiCl\textsubscript{4} were maintained at 18 and 5 sccm, respectively. The chamber pressure during etching was 0.6 Pa. The ICP power was set at 200 W. The flow rate of Cl\textsubscript{2} and the bias power were modified to optimize the etching condition. The different etching conditions were shown in Table 1. To verify the results of the dry etching, etched surface morphology and etching anisotropy were examined by a JEOL JSM-6060 SEM.

**Table 4-1: The ICP etching conditions**

<table>
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<tr>
<th>Condition</th>
<th>Press (Pa)</th>
<th>$P_{\text{ICP}}$ (W)</th>
<th>$P_{\text{Bios}}$ (W)</th>
<th>Cl\textsubscript{2} (sccm)</th>
<th>Ar (sccm)</th>
<th>SiCl\textsubscript{4} (sccm)</th>
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<td>I</td>
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<td>80</td>
<td>30</td>
<td>30</td>
<td>18</td>
<td>5</td>
</tr>
<tr>
<td>II</td>
<td>0.6</td>
<td>200</td>
<td>45</td>
<td>15</td>
<td>18</td>
<td>5</td>
</tr>
<tr>
<td>III</td>
<td>0.6</td>
<td>300</td>
<td>55</td>
<td>15</td>
<td>18</td>
<td>5</td>
</tr>
<tr>
<td>IV</td>
<td>0.6</td>
<td>300</td>
<td>250</td>
<td>15</td>
<td>18</td>
<td>5</td>
</tr>
<tr>
<td>V</td>
<td>0.6</td>
<td>80</td>
<td>15</td>
<td>30</td>
<td>18</td>
<td>5</td>
</tr>
</tbody>
</table>

After the ICP etching, the ELO-GaN was put into MOCVD chamber for the third time to grow the GaN/InGaN multiple quantum wells (MQWs) and GaN cap layer on it. Veeco DI 3100 AFM was used to investigate the surface morphology. After the growth of MQWs and GaN cap, the top SiO\textsubscript{2}/SiN\textsubscript{x} DBR were grown by PECVD. The cross section of the structure was observed by SEM examination.
Figure 4.8: The SEM cross-section image of ELO-GaN grown with pulse NH$_3$ treatment.

Figure 1 (a) shows the SEM cross-section image of ELO-GaN growth. The thickness is about 11 μm. It is clear that with pulse NH$_3$ treatment in ELO-GaN growth, the tilts were greatly reduced and ELO-GaN stripe is perfect rectangular in cross-section. It gives a very good agreement of the top DBR and the bottom DBR for VCSELs and polariton lasers.

The ICP with Cl$_2$/Ar/SiCl$_4$ gas mixtures was used for the etching of ELO-GaN patterns. The GaN etching process produces chemical byproducts such as GaCl$_3$ which is volatile and was pumped out. Smooth surface after dry etch of ELO-GaN is essential to subsequent growth of good quality of device structure. This requires not only overcoming the strong nitride bond energy but also adjusting the process conditions to deal with inherent defects in window region of ELO-GaN. While the defects in the window region appear to be particularly sensitive to etching conditions and respond by etching faster or slower than the surrounding wing region of ELO-GaN. Unacceptable damaged surface or surface morphologies with pits or peaks formation will result if the balance between the physical and chemical aspects of the process is not maintained$^{[15]}$. So in order to get good surface, it is needed to balance the physical and chemical components during ICP etching.
While such balance appear some kind of sensitive. Another thing is the edge effect during the etching of the ELO-GaN patterns. An un-optimized etching process can also result in the peaks or facets near the edge.

Figure 4.9: (a) cross-section SEM image and (b) top-view SEM image of ELO-GaN after etching with pyramids with etching condition I.

First, we tried ICP etch parameter same as what we did for ~400 nm mesa etch in LEDs and HFETs fabrication, which is shown in condition I in Table 1. The total etching thickness of ELO-GaN was around ~6 μm. Figure 2 are the images of etched ELO-GaN patterns with two kind of un-optimized etching process for reference. Fig. 2 shows the cross-section and top-view SEM image of ELO-GaN after etching with pyramids along the stripes. Such pyramids seem large when compared with the size of ELO pattern. The reason of the formation of the pyramids maybe that the chemical component of the etching is much lower than the physical component. Moreover, we have to notice that the dielectric DBRs were also etched during relatively low power ICP etch.

Then the bias power was increased to 200/45 mW and the flow rate of Cl₂ was decreased to 15 sccm to enhance the physical component of the etching to balance the chemical one. Figure 4.10 shows the SEM image of ELO-GaN after etching with condition II in Table 1.
Figure 4.10: (a) cross-section SEM image and (b) top-view SEM image of ELO-GaN after etching with pyramids with etching condition II.

It can be seen that there was no pyramids in the window region which means that the enhanced physical component successfully removed pyramids which were generated during low power etch. Both the flat window and wing regions can be obtained with this etching condition. And the whole pattern region looks like very smooth. However, the peaks near the edge region appeared after etching. We believe that the formation of peaks near the edge corresponds to that the dielectric material was sputtered during the etching. Since we already observed that dielectric DBRs were etched in low power ICP etch with condition I, the etching rate of dielectric should be increased with higher ICP power. The etched dielectric material was sputtered during etching, and the sputtered dielectric material deposited again near the edge of ELO-GaN pattern. Such deposited dielectric material near the edge will act as the masks in the subsequent etching of GaN. In this way the peaks become larger and larger.
Figure 4.11: Cross-section SEM image I and (b) cross-section SEM image II of ELO-GaN after etching with pyramids with etching condition III.

When the physical component in the etching dominates more largely, the sputtered dielectric material cannot deposit near the edge, which is shown in Figure 4.11 with etch condition III. It is clear that the peaks at edge disappeared when we further increased etching power. However, non-uniform etching is observed across the ELO-GaN layer, where the center etching rate is relatively higher than the edge. This non-flat ELO-GaN surface is an obstacle for the following cavity growth.

Figure 4.12: Cross-section SEM image and (b) top-view SEM image of ELO-GaN after etching with pyramids with etching condition IV.

To overcome the non-uniform etching rate, we further increased the ICP and bias power to 300/250 W. From Figure 4.12 (a), we can see that after long time etching, there are no pyramids or peaks observed on sample surface. Moreover, the ELO-GaN surface is
relatively smooth. The DBRs without ELO-GaN covering were completed etched away, and the bottom GaN beneath them was also removed. The edge of ELO-GaN wing is very sharp, indicative of high physical component in ICP etching. In Figure 4.12 (b), we can see low density of pits appeared in the window region of ELO-GaN. Unlike the wing region of ELO-GaN, which is nearly dislocations free, the window region of ELO-GaN is with high density of dislocations. During high power ICP etch, pits appeared due to the high dislocations density. However, it would not affect our cavity design, since the cavity would be deposited only in wing region of ELO-GaN.

After the ELO-GaN was etched to around 400 nm (the center of the pie-shaped sample) and the peaks near the edge were removed, InGaN active region and $p$-GaN layer were grown on the etched ELO-GaN. The thickness of the structure between top and bottom DBR is designed to be around 1.5~2 $\mu$m. This structure is hopeful for the application in the VCSEL structure.

### 4.2.2.3 Full cavity VCSELs

After reducing the ELO-GaN thickness down to 400 nm using ICP etching, the sample was loaded into the MOCVD chamber and subjected to an in situ H$_2$ treatment at high temperature ($950 ^\circ$C) to remove the residual etching damage on ELO-GaN surface. Since multi-DHs have exhibited a superior approach for quantum efficiency enhancement compared to MQW as LED active regions, multi-DH active regions were employed in VCSEL cavity, shown in Figure 4.13 (a). The cavity active region features 2 periods of hexa 3 nm-thick In$_{0.13}$GaN DH (separated by 3 nm-thick In$_{0.01}$GaN barriers) separated by 160 nm ($\lambda$) In$_{0.01}$GaN underlying layer. The designed emission wavelength was 410 nm.
Here two periods of hexa 3 nm In$_{0.15}$GaN DHs were employed in order to feed to cavity more photons to obtain better cavity mode. In addition to vertical cavity sample a test sample was simultaneously grown on c-plane GaN on sapphire substrate in order to compare the optical quality of the vertical cavity active region. Similar pulsed-NH$_3$ method as ELO-GaN growth was employed for p-GaN growth to ensure p-GaN would spread SiO$_2$ layer, where the p-contact would be formed. The full vertical cavity was finished with 13.5 pair SiO$_2$/SiN$_x$ top DBRs deposited using PECVD system with target stop band wavelength around 400 nm.

![Diagram of VCSEL full vertical cavity structure on c-sapphire](image)

**Figure 4.13:** (a) the VCSEL full vertical cavity structure on c-sapphire. (b) Electric field inside the cavity with respect to distance where an InGaN active region is placed at the antinode of the electric field inside the cavity.

It should be noted that a 300 nm-thick SiO$_2$ layer was deposited using PECVD system followed with a photolithography procedure after n-GaN growth. 300 nm SiO$_2$ layer was deposited on ELO-GaN layer after n-GaN growth. Then photolithography pattern was put
on the oxide layer, followed by buffer oxide etcher (BOE) wet etching. After etching, 8 um wide SiO$_2$ stripes separated by 10 um were achieved with the same orientation as ELO stripes. The window regions of ELO-GaN and separation region between ELO-GaN stripes were covered with SiO$_2$ and will not participate in current injections. Therefore, the SiO$_2$ layer ensures the electrical injection goes through InGaN active region grown on dislocation free ELO-GaN wing region only. Figure 4.13 (b) demonstrates the multi-DH active regions are designed to place on electric field antinodes in the cavity in order to maximize the cavity emission. However, due to the thickness variation of the ELO-GaN layers beneath the active regions, the placement of the active regions within the cavity varies across the sample. Further optimization to form homogeneous ELO GaN layers is necessary for better optimized vertical cavity structures.

### 4.2.3 Optical characterization for VCSEL

The VCSEL full cavity structure was characterized using reflectivity (Xe lamp source) for the reflectivity spectrum in the full cavity structure, shown in Figure 4.14. It is clear that 99% reflectivity is achieved for the top SiO$_2$/SiNx DBR with a stop-band width of $\sim$80 nm, which is much larger compared with that of semiconductor AlN/GaN DBRs ($\sim$18 nm). The full-cavity reflectivity spectrum showed clear dips corresponding to the cavity modes at $\sim$ 400 nm and $\sim$ 412 nm.
Figure 4.14: Reflectivity spectrum of the full cavity structure. Cavity modes are observed around 400 and 412 nm can be clearly seen in inset.

Micro-PL measurement was employed to get the Q factor of the VCSEL sample as well as the simultaneously grown reference sample. The samples were excited normal to the surface from the top DBRs side using a HeCd laser with wavelength 325 nm. The 325 nm wavelength excitation ensured to excite the active region outside of the top DBRs stop band from 350 nm to 430 nm (see Figure 4.14). The emission was also collected normal to the surface from the top DBR side. The excited area was in the order of ~ 2 μm in diameter. Figure 4.15 shows the PL spectra for the full cavity structure, half cavity structure and reference sample. The half cavity Q-factor is 60, 3 times higher than reference sample Q-factor 20, clearly showing the confinement of PL spectrum from bottom DBRs. The micro-PL for the cavity mode has an average Q-factor of 500 across
the wafer, which was substantially higher compared with half cavity Q-factor after the top DBRs deposition. The result proves the obtained high quality full cavity structure, which is very promising for the electrical operation. However, we have to note that the vertical cavity thickness varies across the wafer. Owing to non-uniform vertical thickness of ELO-GaN after MOCVD growth, during ICP-etching on ELO-GaN, some portion of ELO-GaN stripes remained thickness of a few micro while some portion of ELO-GaN stripe was nearly etched away.

![Graph showing PL spectra](image)

**Figure 4.15:** PL spectra for the full cavity structure (red), half cavity structure (black) and reference sample (blue).

### 4.2.4 Fabrication and electrical characterization for VCSEL

Fabrication was processed on the full cavity for electrical characterization. One challenge for the GaN-based VCSEL fabrication with both dielectric DBRs is current confinement. Since the ELO-GaN window region has a high density of dislocations, which could be
current leak path. It is necessary to do a selective oxidation to prevent current spreading to window region. 300 nm SiO₂ layer was deposited after n-GaN growth followed with photolithography pattern. 8 um wide SiO₂ stripes separated by 10 um were achieved with the same orientation as ELO stripes to cover SiO₂ layer on the window regions of ELO-GaN and separations between ELO-GaN stripes. Therefore, only InGaN active region grown on high quality ELO-GaN wing region will participate in current injections. To form the n-contact, the top DBRs and the whole active regions were etched away by ICP dry etching. Ti/Al/Ni/Au (30/100/40/50 nm) were served as n-contact, followed with RTA annealing at 860°C for 1min. Pulse-NH₃ method was also employed for p-GaN growth to ensure p-GaN would spread SiO₂ layer, where the p-contact would be formed. Ni/Au (5/5 nm) was served as p-contact with Ni/Au (40/50 nm) served as p-pad for probes. The full schematic of fabricated devices is shown in Figure 4.16.

![Figure 4.16: Cross-sectional schematics of fabricated VCSEL device with electrical contacts.](image)

The mask designs to produce the vertical cavity devices with different sizes using photolithography and e-beam deposition techniques are shown in Figure 4.17. Figure 4.17 (a) illustrates the mesa patterns on ELO-GaN stripes, while Figure 4.17 (b) shows...
the final device shapes with contact layers. The devices are prepared to constitute on changing mask patterns in the dimensions of 540x540, 200x200, 20x40, 40x20, 20x20, 20x10 \( \mu \text{m}^2 \). The \( p \)-contacts are shown in black stripes, while \( p \)-pad (red net) was deposited on top of dielectric DBRs. \( N \)-contacts (Ti/Al/Ni/Au) deposited on regions between mesa patterns.

Figure 4.17: VCSEL devices on the mesa, (b) the final device shapes with contact layers.
Chapter 5  Conclusions and future research

5.1 Conclusions

InGaN based LEDs have been widely used in displays of TV, computers, and cell phones. And more applications have been focused on the general lighting market due to their high efficiency and long lifetime. However, high power LEDs suffer efficiency droop at high injection levels. The theoretical and experimental results along with work done by other researchers have ruled out the possibility of Auger recombination in wide band gap GaN based LEDs. We have proposed that the poor hole transport in GaN is an obvious contributing factor for such efficiency droop. Delta-doping with Mg of barriers significantly improves the quantum efficiency of LEDs with active regions composed of hexa 3-nm DHs. With careful design of the active regions, the significant improvement in hole injection was achieved without degradation of the material quality due to the presence of Mg. With Mg delta-doping of the first barrier on the LED n-side, the relative peak EQE improves by 20% in compared with the reference structure. Doping of the first and second barriers on the n-side does not result in further improvement of relative EQE compared to the case of one doped barrier. This possibly indicates that further improvement in hole injection is canceled by the reduced active region quality due to higher Mg-doping. The both LED structures with Mg delta-doped barriers effectively suppress the efficiency droop at high injection level compared to the counterpart with undoped barriers.

Armed higher quantum efficiency in our LED structures, we investigated various multi-DH active region designs to maximize the light output. By using multiple thin DH
structures, the relative EL efficiency was enhanced greatly by increasing number of DH up to hexa 3 nm DH. It is clear that with more DHs in the active region (less than 6), the LED can handle more injection power and thus favors high power applications. Our optimization of SEI structures increasing SEI thickness from 5 nm to 30 nm per step further improved relative EL efficiency for single 3 nm DH and move the peak efficiency of 3 nm DH to the comparable level with quad 3 nm DH albeit single 3 nm DH shows ~2 time lower EL efficiency than quad 3 nm DH at high injection levels. It is shown that a good design of SEI may play an important role for the efficiency improvement of LEDs, especially for LEDs with thin active layers.

Novel pulsed-NH₃ growth techniques have been used for GaN-based VCSEL structure with growing active layers on nearly defect free epitaxial lateral overgrown (ELO) GaN layers embedded in bottom and top dielectric DBRs. ELO-GaN growth provides a superior solution to VCSELs. While VCSEL on bottom AlN/GaN DBRs suffers narrow stop-band and high density of defects, resulting quality degradation of active layers, ELO-GaN growth technique could provide wider stop-band dielectric DBRs and high quality InGaN active regions. Quality factors up to 1300 was obtained for the VCSEL structure with dual hexa 3 nm DH active region on semiconductor bottom DBRs. In order to achieve the appropriate ELO-GaN for vertical cavity structure, MOCVD growth parameters was studied for higher lateral to vertical (L/V) growth ratio for ELO-GaN wings. Furthermore, the etching parameters to thin down ELO-GaN using inductively coupled plasma (ICP) technique was investigated to obtain desired vertical cavity lengths and reduce defects density after etching. Final cavity structure was obtained after the lithography techniques used following with e-beam deposition for the electrical contacts.
5.2 Future research

From aforementioned discussions supported by massive theoretical and experimental results, we can conclude that in order to further mitigate efficiency droop and improve the GaN-based LEDs, hole concentrations must be further increased. However, standard GaN growth conditions by MOCVD lead to hole concentration typically on the order of $4.7 \times 10^{17} \text{ cm}^{-3}$ since the Mg activation energy in GaN is high and the formation of the Mg-H complexes inhibit the ionization of Mg acceptors. Using higher growth pressure and lower temperature can enable higher hole concentrations. However, low growth temperature leads to GaN material quality degradation. In addition, this approach is limited by self-compensation attributed to nitrogen vacancy complexes in $p$-GaN. Due to the motion of Fermi level, nitrogen vacancies are expected to have a major impact on $p$-GaN. However, with higher NH$_3$ partial pressures during growth and careful post annealing, the nitrogen vacancy density can be minimized. For $p$-GaN growth with Ga polarity, the Mg incorporation can induce the stacking faults from GaNGaN to GaNMgNGa, inverting the GaN polarity from Ga-face to N-face. As the growth proceeds, additional Mg atoms migrate towards these stacking faults leading them to develop along several inclined planes, and eventually form pyramidal-shape defects, called pyramidal inversion domains. Delta-doping of Mg into GaN was found to be virtually free of such inversion domains extended defects due to the hindering of the vertical diffusion of Mg inhibited by GaN interlayers, which in turn improved the surface morphology. It was also reported that Mg delta-doping improves not only the $p$-type GaN conduction, but also significantly suppresses the dislocation densities, which is
beneficial to the leakage current and lifetime of LEDs.\textsuperscript{95} It has also been demonstrated that incorporating Mg-doped AlGaN/GaN superlattice structure into devices could enhance the hole conduction in the lateral direction. However, the enhancement of hole conduction in the vertical direction by employing a superlattice structure is limited because a superlattice structure simultaneously introduces potential barriers for hole conduction in the vertical direction.

![Figure 5.1: delta-doping profile implementation: Stage I: undoped GaN growth (u-GaN); II: Nitridation; III: Mg incorporation (constant Mg molar flow for all the samples);](image)

Using the Mg δ- doping technique, hole concentration as high as $10^{18}$ cm$^{-3}$ has been achieved by MOCVD growth method\textsuperscript{96}. We expect that, by tuning the growth parameters (including growth temperature, Mg flow rate, interruption time and undoped GaN spacer layer thickness) during growth of Mg δ- doped GaN, the solubility limit of Mg into GaN can be affected in a controlled way, allowing to enhance the incorporation of Mg ions, to hinder the self-compensation mechanisms and therefore improve the concentration of active carriers in the layers. For $p$-type doping studies, a p-$n^{-}$-$n^{+}$ layer structure was grown. The growth conditions for top Mg δ- doping GaN have been varied by changing the undoped GaN spacer thickness from 5 nm, 7.5 nm, 10 nm, to 12.5 nm. The bottom n
type layers were Si-doped with carrier concentration around $3 \times 10^{18}$ cm$^{-3}$. This was capped with lightly Si doped GaN ($5 \times 10^{17}$ cm$^{-3}$) layer of 200 nm thickness, and finally 500 nm Mg δ- doping GaN. This structure is useful for making electrical measurements on Mg doped layer because the resulting p-n junction has a high reverse breakdown voltage; as a consequence a high bias can be applied between two $p$-contacts and only hole current will flow between them, enabling accurate measurements. In addition, by forming p-n junction devices, we can test the I-V characteristics for the p-n junctions simultaneously. The results are shown in Table 5-1 using TLM measurement [Figure 5.2 (a)] assuming hole mobility 5 cm$^2$/Vs.

<table>
<thead>
<tr>
<th>Mg delta doping</th>
<th>Specific contact resistance (Ohm cm$^2$)</th>
<th>Sheet resistance (Ohm)</th>
<th>Hole concentration (cm$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(5nm)</td>
<td>$3.5 \times 10^{-1}$</td>
<td>$1.2 \times 10^{6}$</td>
<td>$0.2 \times 10^{17}$</td>
</tr>
<tr>
<td>(7.5nm)</td>
<td>$1.6 \times 10^{-2}$</td>
<td>$1.6 \times 10^{3}$</td>
<td>$1.5 \times 10^{17}$</td>
</tr>
<tr>
<td>(10nm)</td>
<td>$6.6 \times 10^{-3}$</td>
<td>$6.3 \times 10^{4}$</td>
<td>$4.0 \times 10^{17}$</td>
</tr>
<tr>
<td>(10nm)</td>
<td>$6.2 \times 10^{-3}$</td>
<td>$6.5 \times 10^{4}$</td>
<td>$3.8 \times 10^{17}$</td>
</tr>
<tr>
<td>Mg 200sccm uniform</td>
<td>$4.8 \times 10^{-3}$</td>
<td>$3.3 \times 10^{4}$</td>
<td>$7.6 \times 10^{17}$</td>
</tr>
</tbody>
</table>
Figure 5.2: (a) Cross-sectional schematic of TLM pattern; (b) Measured hole concentration as a function of u-GaN spacer thickness
As presented in Error! Reference source not found. (b), with increasing u-GaN spacer thickness up to 10 nm, the hole concentrations continue improving. The highest hole concentration obtained so far is \(4 \times 10^{17} \text{ cm}^{-3}\) with u-GaN thickness. Further increasing u-GaN thickness results in reduced hole concentration probably due to decreased Mg doping concentration. Although the obtained hole concentration is still less than our standard continuous Mg doping \(p\)-type GaN with hole concentration \(7 \times 10^{17} \text{ cm}^{-3}\), the Mg delta-doping GaN optimization is still on the way. Further optimizations are required in order to achieve higher hole concentration including (1) optimizing Mg source flow; (2) optimizing u-GaN spacer growth V/III ratio.
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EDUCATION

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Electrical Engineering, Ph.D., December 2014

Chinese Academy of Science, Beijing, China
Material Science and Engineering, M.S., July 2008

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Microelectronics, B.S., July 2005

TECHNICAL SKILLS

- Semiconductor device fabrication and processing: Photolithography/Wet Etching /Metallization/E-beam Deposition/Sputtering/Plasma Etching/Wire Bonding
- MOCVD epitaxial thin film growth
- PECVD deposition of SiO$_2$/SiNx
- Device testing: C-V, I-V, AC/DC/RF, LCR meter, TLM, EL, integrating sphere
- Characterizations: XRD, SEM, AFM, PL, Profilometry, Hall measurements
- Computers: Matlab, JMP, Silvaco TCAD, Mathematica, Origin, MSOffice

PROFESSIONAL EXPERIENCE

Microelectronics Materials and Device Laboratory--Virginia Commonwealth University

- Research Assistant, Aug. 2009 to Present
  - Designed InGaN active regions (single and multiple quantum wells) to improve quantum efficiency of LEDs
  - Optimized electron cooler layers for efficiency droop mitigation to enhance LEDs light-output
- Designed novel delta p-doped quantum barrier in InGaN active regions and optimized p-GaN structure to improve hole injection of LEDs
- Simulated LED devices using Silvaco TCAD and incorporated into structure designs
- Developed novel growth technique for ELO-GaN structures on SiO₂/SiNx Distributed Bragg Reflectors (DBR) on sapphire
- Achieved crack-free semiconductor DBRs with reflectivity as high as 98% for vertical cavity laser (VCSEL) applications
- Developed InGaN/GaN phonon cavity on AlN/GaN superlattices
- Investigated dislocations, point defects, basal and prismatic stacking faults of III-Nitride thin film layer
- Developed dry etching process to achieve smooth surface with minimized surface damages and defects density
- Demonstrated technology to obtain high quality semi-polar and non-polar GaN layers on Si substrates
- Improved HEMTs channel mobility and reduced sheet resistance
- Developed novel couple/dual-channel HEMTs to reduce hot phonon effect and improve devices reliability
- Simulated HEMT structures in Silvaco TCAD and incorporated into growth designs
- Developed dielectric thin films for passivation to improve HEMTs performance
- Optimized plasma etching conditions and eliminated HEMT device leakage current
- Reduced specific ohmic contact resistance by one order of magnitude for HEMTs
- Investigated AlGaN HEMT on Si substrates

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KEY PUBLICATIONS


