Theoretical study of segregation kinetics of indium in indium gallium nitride and magnesium in magnesium-gallium nitride grown by molecular beam epitaxy

Irena Vidhya Mabel Stanley
University of Nevada, Las Vegas

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THEORETICAL STUDY OF SEGREGATION KINETICS OF In IN InGaN AND Mg IN Mg-GaN GROWN BY MOLECULAR BEAM EPITAXY

by

Irena Vidhya Mabel Stanley

Bachelor of Science
University of Madras, India
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A thesis submitted in partial fulfillment
of the requirements for the

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Irena Vidhya Mabel Stanley

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Master of Science

Examination Committee Chair

Dean of the Graduate College

Examination Committee Member

Examination Committee Member

Graduate College Faculty Representative
ABSTRACT

Theoretical Study of Segregation Kinetics of In in InGaN and Mg in Mg-GaN Grown by Molecular Beam Epitaxy

by

Irena Vidhya Mabel Stanley

Dr. Rama Venkat, Examination Committee Chair
Professor of Electrical and Computer Engineering
University of Nevada, Las Vegas

A rate equation model including all physically relevant surface processes is developed for the study of In segregation in InGaN and Mg segregation in Mg-GaN MBE growth. In InGaN growth, the simulations were carried for a variety of growth conditions spanning the growth parameter space: substrate temperature in the range of 500-700 °C; Ga flux in the range of 1.17-8.98 nm/min; In flux in the range of 0.39-28.74 nm/min and N flux in the range of 4.7-12 nm/min. Results of In incorporation obtained from simulations are within 1% agreement with the experiments reported in the literature. In segregation is found to be negligible below 580 °C. Above 640 °C, the segregation dominates the kinetics. This temperature dependence is found to be independent of the fluxes. In Mg-GaN growth, simulations were carried for various growth temperatures in the range of 600-750 °C with constant flux rates of Mg, Ga and N. For the given flux rates, it is found that Mg segregates to the surface with the increase in temperature. Above 750 °C a dopant depleted zone is formed below the surface layer. Results obtained from simulations are in good qualitative agreement with the experimental data.

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CHAPTER 1

INTRODUCTION

Molecular Beam Epitaxy is the leading technique in growing high purity and atomistically controlled hetero-epitaxial layers [1]. The process involves the crystallization of compounds on the substrate by the process of condensation and reaction of vapor in ultra high vacuum. The inner surfaces of MBE growth chamber are very clean as the chamber is evacuated to high vacuum level up to \(10^{-10}\) torr. The high purity heterostructures for device applications of controlled composition and thickness can be grown by MBE due to low growth rates of the order of 1\(\mu m/hr\). Since the molecules in the beam don’t collide with each other before they reach the substrate surface, multi-layered structures with abrupt interfaces and compositional changes can be grown.

The in-situ monitoring techniques such as Auger Spectroscopy and Reflection High Energy Electron Diffraction (RHEED) available in MBE systems make it a highly efficient process tool. The growing surface is monitored as it grows and the composition, thickness, crystallinity and roughness can be dynamically controlled. The precise control of beam fluxes, growth conditions and the in-situ monitoring makes MBE a versatile growth technique, which is more reliable than all others. MBE technique has been used successfully in growing a variety of electronic and opto-electronic devices involving a variety of IV, III-V and II-VI compounds and their alloys. The materials used extensively are \(GaAlAs\), \(InGaP\), \(HgCdTe\) and \(GaAlN\).
Highly efficient light emitting diodes and lasers are fabricated using III-nitride semiconductors [2]. The reason for their prominence in blue lasers and LEDs is their large band gap. In addition, using various III-nitride compounds with bandgap spanning the whole of visible spectrum, opto-electronic devices in all three primary colors can be fabricated. Group III-nitrides are the choice for the opto-electronic devices due to higher device stability for high temperature and high break down fields than the II-VI ZnSe based devices. The recent successful realization of semiconductor light emitting diodes and lasers commercially has paved way for increased interest in thin films wide band gap III-N.

In spite of the increased interest and successful device fabrication [3-20], the precise understanding of the atomistic details of the growth process is lacking. Such details are necessary for reproducing the hetero-structures with abrupt interfaces and varying compositions. Attempts at theoretical modeling of growth included Stochastic models [21, 22], Monte Carlo simulations [23, 24], Quasi thermodynamic models [25, 26, 27] and Rate equation models [28, 29, 30]. Each approach has some advantages and limitations compared to the other. Details of these are presented in Section 2, Literature Survey.

Chen et. al. [31] and Averbeck et. al. [32] investigated the growth kinetics of $\text{InGaN}$ and its relation to growth conditions such as temperature and fluxes. In both their work, it was observed that In segregates to surface at high temperatures independent of the fluxes and flux ratios due to the weaker bond strength of $\text{InN}$ compared to $\text{GaN}$.

Guha et. al. [33], Myoung et. al. [34] and Cheng et. al. [35, 36] studied the mechanism of Magnesium incorporation and segregation in $\text{GaN}$ grown by MBE. The
effect of flux ratio and substrate temperature was investigated and it was observed that $Mg$ segregates to the surface through inter layer migration due to the rapid diffusion of $Mg$ and the dependence of its diffusion rate on substrate temperature and incident flux rate.

In spite the experimental work presented in Ref. [31, 32, 33, 34, 35], a clear understanding of the segregation phenomena is lacking. In this thesis work, the rate equation model developed and used by Fu et. al. [37] is adopted and modified to study the segregation phenomenon and the underlying physics and chemistry. Segregation of $In$ in $InGaN$ and $Mg$ in $GaN$ are studied in their relation to the growth conditions. The models simulated in this work can be developed as software and can be used in the industry, through which the overall cost of crystal production can be minimized.

1.1 Organization Of The Thesis

In Chapter 2, the literature review on MBE, group III-nitrides, $InGaN$, doping issues, models and devices are presented. In chapter 3, a detailed rate equation model including all the appropriate surface processes, is developed for the theoretical study of growth of $InGaN$ and $Mg$ doped $GaN$ by molecular beam epitaxy. Also the computational details and model parameters are presented in chapter 3. The results and discussions are presented in chapter 4. Conclusions are in chapter 5.
CHAPTER 2

LITERATURE REVIEW

2.1 Molecular Beam Epitaxy

The Molecular Beam Epitaxy (MBE) systems can be physically divided into three zones [1]. Each of these zones plays an important role in the growth of an epilayer and any alterations in these zones will seriously impact the growth.

In the first zone the generation of molecular beam is accomplished. The molecular beams are generated from Knudsen effusion cells (K-cells) under ultra high vacuum of the order of $10^{-9}$ torr. Flux from the K-cell is controlled by the shutters situated at the mouth of the cell, which is electronically controlled from outside. The desired composition in the epitaxial film can be obtained by appropriately opening and closing the shutters of particular K-cells and by choosing the particular substrate temperature. The spatial uniformity of the molecular fluxes from the K-cells controls the spatial variation of the thickness on the wafer. The effusion cells are made in various sizes and shapes according to the needs of the technology.

The second zone is the vacuum area through which the molecular beam travels and reaches the substrate. The mean free paths of the molecules are long such that they do not collide during their flight from the K-cell to the substrate. Thus, the molecules do not exhibit viscous flow. At the periphery of this zone, the in-situ growth monitoring tools
such as Auger Spectroscopy and Reflection High Energy Electron Diffraction (RHEED) are also situated.

The third zone is the area where the crystallization occurs on the substrate due to interaction of molecular beams at the substrate temperature. Various surface processes take place in this area that result in the growth of the epilayer. The surface processes are: adsorption, surface migration and dissociation of the adsorbed molecules, incorporation and thermal desorption. The adsorption is of two types. One is physisorption in which the adsorbate is bonded to the substrate through weak bonds (Vander waals force) and no electron transfer between the atoms is involved. The other is chemisorption in which the adsorbate and substrate chemically react and transfer of electrons between the atoms is involved. Van der Waal bonding is of the order of 0.1-0.2 eV whereas the chemical bonding is typically 1-2 eV.

The in-situ monitoring is done by RHEED (Reflection High Energy Electron Diffraction), REMS (Reflection Mass Spectrometry), MBMS (Modulated Beam Spectrometry), and RDS (Reflectance Difference Spectroscopy). RHEED is used to monitor the crystal structure and the microstructure of the growing surfaces. REMS and MBMS are used to monitor the surface chemistry and composition on the growing surface. RDS is used to monitor the composition through the optical properties of the growing surfaces.

Ellipsometry and laser interferometry are other optional techniques that are used. Since these in-situ techniques do not interfere with the flow of molecular flux or beams, they are non destructive and provide the direct measurement of the surface structure. In modern MBE systems, several additional surface monitoring techniques are included.
They are: Auger electron spectroscopy (AES), Secondary Ion Mass Spectroscopy (SIMS), X-ray Photon Spectroscopy (XPS), Ultraviolet Photoelectron Spectroscopy (UPS) and Scanning Electron Microscopy (SEM).

AES is used for determining the chemical composition of the growing layer and to determine the bulk chemical composition of the grown structures in terms of depth profiling. SIMS is used for determining the chemical composition of the surface atomic layers and it has high mass resolution. XPS and UPS are used for studying the electronic structure or the energy band distribution at the hetero interfaces. SEM is used for the display of the structure of the growing film or the substrate.

The present day MBE techniques use both Physical Deposition Techniques (PDT) and Chemical deposition techniques (CDT). In PDT, the source material is vaporized at very high temperatures and is transported towards the substrate through a reactor, which is maintained at high vacuum, in the form of vapor without any chemical change. In CDT, the volatile source material is initially produced inside or outside the reactor and is allowed to pass through the reactor to the substrate in the form of vapor. The constituents of the vapor undergo chemical reaction to produce the reactants that aid in the film growth.

2.2 Group III-Nitrides

“Nitrides” is the collective term used for a group of extremely interesting compound semiconductor materials containing N such as InN, GaN, AlN and their associated compounds. These compounds are receiving considerable attention because of their opto-electronic application spanning the entire visible spectrum because these materials and
their alloys have energy band gaps covering the range of 1.9-6.2eV, suitable for band-to-band light generation with colors ranging from red to ultraviolet wavelengths [2].

Group III nitrides occur in both zinc blende and wurzitic structure. The wurzitic structure is the most common in GaN and its related compounds [2]. The research on group III-nitrides is mainly concerned with the aspects such as achieving cubic $\beta$-GaN, using usual techniques to grow GaN at low temperatures, applying AlN buffer layer between the substrate and the film, and using electron beams to achieve $p$-type material. A suitable high quality lattice matched substrate is difficult to obtain in the case of $\beta$-cubic GaN.

Sapphire substrates are used for the growth of GaN in spite of the lattice mismatch between the epilayer and the substrate. The grown layers usually show $n$-type conductivity even though they are not intentionally doped. The donors are probably the residual oxygen or nitrogen vacancies. Use of buffer layers between the epilayer and the substrate helps to improve the surface morphology. AlN is the commonly used buffer layer. Use of GaN buffer layers instead of AlN results in higher quality films [38].

Cheng et. al. [39] demonstrated the growth of GaN films on GaAs surfaces using modified MBE method in which the active nitrogen was supplied from an RF activated plasma source. The GaN films grown on (001), (111) A and (111) B GaAs substrates at 700 °C and under similar conditions were compared. Using photoluminescence (PL) spectra it was shown that the best optimum growth was obtained on (111) B GaAs. Paisley et. al. [40] demonstrated the growth of cubic GaN on Si by modified MBE. A standard effusive cell was used for Ga and a microwave glow discharge was used to activate nitrogen prior to deposition. The grown films were analyzed using Auger
Electron Spectroscopy and Transmission Electron Spectroscopy and were found to be nominally stoichiometric and zinc blende crystal.

Lie et al. [41] studied the epitaxial growth of zinc blende and wurzitic GaN films on silicon substrate by Electron Cyclotron Resonance (ECR) assisted MBE, using a two-step process. Initially a GaN buffer layer was deposited at low temperature followed by the growth of thicker layers of GaN at higher temperature. GaN films were found to have either zinc blende or wurzitic structure depending on the single crystalline or polycrystalline buffer layers used, respectively. The growth of GaN by ECR assisted MBE was studied by many researchers [42, 43, 44, 45, 46]. Molnar et al. [44] studied the importance of ionic species in the growth of GaN by ECR. The ion bombardment was found to have a profound effect on the kinetics of the growth. A transition of film growth from island to three-dimensional mode takes place by varying the microwave power in the ECR’s discharge source.

The growth of GaN by MBE with radio frequency (RF) nitrogen plasma was studied by [47, 48, 49]. Kubo et al. [47] investigated the growth of homoepitaxial growth of GaN thin layer on MOCVD grown GaN substrate. It was shown that the homoepitaxial GaN had higher crystalline quality by the surface and optical measurements that were made on the sample. It was proposed that the problems of hetero interface between GaN and sapphire would be improved by homo epitaxial growth. High quality growth of AlN layers on Si substrates by Plasma assisted MBE was studied by Sanchez-Garcia et al. [48]. The growth conditions were optimized in order to obtain AlN as buffer layers in GaN film growth. It was shown that the best films were obtained at substrate temperature greater than 900 °C and the V/III flux ratios close to stoichiometric values.
High quality GaN and AlN growth by gas source MBE using ammonia as the nitrogen source was reported by Yang et. al. [50] in 1995. The growth rate as high as 1μm/hr., the highest ever proposed in gas source MBE was achieved. The low temperature photoluminescence of as-grown materials was dominated by band edge emissions, which indicated the high quality of the materials. Ammonia has been used for the growth of GaN by MBE on sapphire substrates by Grandjean et. al. [51]. Their experiments were performed, by varying the V/III ratios in the range of 1 - 4. It was found that the increase in V/III ratio improves the material properties, both in terms of optoelectronic and structural quality.

Growth Kinetics of group III nitrides using modified MBE was studied by Foxon et. al. [52]. A fixed Ga arrival rate of 3×10⁴ atoms/cm²s and constant substrate temperature of 600 °C were used in their experiments to grow GaN films. The final thickness of the film measured by optical interference method was used to deduce the average growth rate. The growth rate was found to increase with optical emission detector output and saturate above 0.7 values, whereas the Ga droplet density formed during the growth showed the reverse characteristic. The film growth under N-rich condition in the temperature range of 400-700 °C was also studied and it was observed that there was no change in the growth rate.

The dependence of growth rate on temperatures and V/III ratio during GaN MBE was studied by Alexeev et. al. [53]. The samples were observed through optical reflectivity measurements. It was found that the GaN desorption becomes observable at temperature above 800 °C which directly relates to reduction in growth rate observed. Desorption was found to be independent of V/III ratio within N-rich conditions. Growth
rate reduction of GaN due to the surface accumulation of Ga was studied by Crawford et. al. [54]. It was observed that at high temperatures, the growth rate decreases due to Ga desorption and decomposition of GaN. At low temperature, the N incorporation efficiency was the rate limiting process.

Zsebok et. al. [55] studied the surface morphology of the GaN as a function of N/Ga ratio grown on GaN by MBE. The different samples grown under N-rich, Ga-rich and stoichiometric conditions were analyzed through the SEM. It was concluded that the defect formation at the hetero-interface depends on the N/Ga flux ratio. Smith et. al. [56] studied the surface reconstruction using the RHEED measurement. It was shown that the smooth (streaky pattern) to rough (spotty pattern) transition of the surface occurs at low Ga/N flux ratio.

Many researchers [57, 58, 59, 60] studied the structural and electronic properties of group III nitrides. The optical properties of the crystal depend on the band structure, confined states of energy levels, polarization defects, lattice mismatch between the active layer and the substrate, dopants and defects in the active layer [38, 60, 61]. Higher bandgap enables the material to withstand higher temperatures due to low intrinsic carrier concentrations. The localized energy states confine the carriers to particular energy levels, increasing the intensity of the light. Lattice mismatch introduces strain in the active layer, which leads to changes in the optical property of the material due to lattice constant and hence bandgap energy changes. Dopants and defects act like a recombination center and results in non-radiative recombination of carriers, which in turn, reduces the intensity of the light.
2.3 \textit{InGaN}

\textit{GaN} does not emit a strong band-to-band emission at room temperature. The non-availability of proper substrate (lattice matched) and the difficulty of \textit{p}-type doping of \textit{GaN} paved way for the alloys and hetero structures involving \textit{In} and \textit{Al}. Indium is added to \textit{GaN} to obtain strong band-to-band emission, as \textit{In} introduces deep localized energy states in the bandgap. The \textit{In} content in the \textit{InGaN} can be changed to realize the strong band-to-band emission from green to UV range \cite{62}. Matsuko \cite{63} observed the dependence of the absorption coefficient on the photon energy of \textit{InGaN} epitaxial layer grown at 500 °C. The plot of absorption co-efficient squared vs. the photon energy in each of the epitaxial layer is linear in the high-energy regime. From their observation \textit{InGaN} has been concluded to have direct band gap as \textit{InN} and \textit{GaN}.

Recently, \textit{InGaN} has become the most promising material for achieving good electrical and optical characteristics from the LEDs \cite{38, 60, 61, 62, 64}. Several researchers have reported growth methods of realizing high quality \textit{InGaN} \cite{31, 32, 65, 66}. Bottcher \textit{et. al.} \cite{67} studied the incorporation behavior of \textit{In} during MBE growth of \textit{InGaN}. The experiments were carried out by varying the \textit{In/Ga} flux ratios and with different film thickness. The samples were analyzed using energy dispersive X-ray microanalysis, high resolution X-ray diffraction and photoluminescence spectroscopy. It was found that \textit{In} incorporation was strongly affected by the \textit{Ga} and \textit{N} fluxes and was limited by excess of nitrogen compared to gallium. It was also found that \textit{In} composition was strongly dependent on the thickness of \textit{In}_{x}Ga_{1-x}N film.

Okamoto \textit{et. al.} \cite{68} investigated the effects of atomic hydrogen irradiation on the \textit{In} incorporation in \textit{InGaN} films grown by RF plasma assisted MBE growth. The
experiments were carried out for varying substrate temperature and $H_2$ flow rate, while $N$, $Ga$ and $In$ flux were kept constant. It was found that the atomic hydrogen irradiation increased the $In$ incorporation as the samples grown with $H$ had higher $In$ content compared to samples without $H$ in the substrate temperature range of 640 – 700 °C.

O’Steen et. al. [69] also studied the effect of V/III flux ratio and substrate temperature on the $In$ incorporation behavior in $In_xGa_{1-x}/GaN$ hetero-structure grown by RF plasma assisted MBE. Two sets of super lattices were grown. For the first set, the substrate temperature was held constant and the incident fluxes were varied. For the second set, the fluxes were kept constant and the temperature was varied over the range of 540-670 °C. Average alloy composition, $In$ loss and $In$ incorporation efficiency was observed as a function of V/III flux ratio and substrate temperature. The nominal layer composition of $In$, i.e., $x$, was 30% in $In_xGa_{1-x}N$. It was found that the results obtained from these experiments were consistent with the $In$ loss that arises during the growth from thermally activated surface segregation and surface desorption processes.

$In$ segregation during the growth of $InGaN$ by plasma assisted MBE, was found by Chen et. al. [31, 70]. The dependence of $In$ incorporation on growth temperature and group III/V ratio was reported. It was found that $In$ incorporation decreases when the growth temperature was increased and decreases when the group III/V flux ratio was increased under metal rich conditions. $In$ incorporation was found to increase with flux ratio under nitrogen rich conditions. The surface morphology was also studied using STM images [31, 70, 71]. It was found that the incorporation decreases with the increase in growth temperature, it decreases with the increase in III/V flux ratio under metal-rich conditions and increases with the flux ratio under nitrogen rich conditions. Indium
surface segregation is caused due to the fact that InN bond is weaker than the GaN bond [31]. A metal atom forms four bonds with nitrogen atoms in the bulk, whereas on the surface a metal atom forms only one bond with the N atom in the N-polar InGaN. So the Ga atoms on the surface switch sites with In atoms in the bulk as it is energetically favorable for them.

Growth of GaN, AlGaN and InGaN for varying growth conditions were reported in [32]. About 40 monolayers of InGaN were grown with different fluxes and temperature between 500-700 °C. It was found that below 500 °C, Ga was strongly bonded and displaced In atoms. Above 640 °C, high quality layers were obtained. When the Ga flux was higher than the In flux, no In was incorporated in the epilayer.

Chen et. al. [65] also observed the spontaneous formation of nano structures in InGaN. They showed that these structures arise due to the strain in the surface layers and the relative weakness of In - N bond compared to the Ga-N bond.

2.4 Doping

The column II impurities when added to GaN can either substitute for Ga to form single acceptors or for N to form deeper triple acceptors. In order to make electronic devices, pn junctions are to be formed. n-type GaN are easily grown but the p-doping of GaN is difficult. The vapor pressure of Mg is very high and it evaporates at around 250 °C. The growth of GaN by MBE is carried around 750 °C, which is much higher than 250 °C. This compatibility in temperatures has prompted many research works for the understanding of the incorporation behavior of volatile Mg into the epitaxial GaN. The thermal activation energy required for ionization of Mg is approximately 200meV. At
room temperatures, only few percent of acceptor atoms ionize and so to achieve the $p$-type conductivity, large concentration of Mg in the layer is required [72].

Amano et. al. [73] was the first in obtaining the $p$-type GaN films using Mg as a dopant. They used MOCVD and post low energy electron beam irradiation (LEEBI) treatment. LEEBI treatment was done after the growth to obtain a low resistivity $p$-type GaN film. Saparin et. al. [74] investigated LEEBI treatment effects on Zn doped GaN. But the researches could not succeed in reproducing the doping. Nakamura et. al. [75] were the one who found that the $p$-type GaN with low resistivity can be obtained by thermal annealing of the MBE grown GaN crystals.

The incorporation behavior of Mg in GaN was studied by [33, 35, 72, 76]. Guha et. al. [33] worked on the samples that were grown by MBE using RF source for nitrogen. Infrared pyrometry and SIMS were used to measure the substrate temperature and the Mg concentrations, respectively. Experiments were carried out in two sets. In the first set, the temperature was kept constant at 750 °C and the Mg fluxes were varied in the range of $0.04-4 \text{ ML/s}$. The other set of experiments were carried out with constant Mg flux and varying temperature. It was observed that the Mg incorporation is invariant to the arriving Mg flux and is strongly dependent on the growth temperature. As the temperature was increased the incorporation of Mg decreased.

Two models were suggested to explain the incorporation of Mg. In the first model [35, 77, 78, 79], it was suggested that there is a surface accumulation layer with dopant incorporation driven by segregation effects. The other model [80] assumed the presence of finite concentration of Mg sites in the growing surface. Cheng et. al. [36] studied the mechanism of Mg incorporation by AES, SIMS and Hall Effect measurements. It was
shown that Mg is uniformly distributed in the bulk and saturates at a concentration of 
\((2-3) \times 10^{19} \text{ cm}^{-3}\). The concentration of Mg was found to decrease from its surface value 
to its bulk value over depth of approximately 10nm, which confirmed that the Mg 
segregates to the surface.

A series of Mg step doped epitaxial GaN layers were grown by Ptak et. al. [72] and 
the incorporation was studied for (0001) or Ga Polarity and (0001) or N polarity. SIMS 
was used to determine the Mg concentration. It was found that Mg incorporation depends 
on polarity and the measurements also supported the surface accumulation of Mg during 
growth. Mg incorporation was found to increase in the presence of atomic hydrogen. 
Myers et. al. [76] studied the doping of GaN with Mg and Be. It was also observed that 
the Mg incorporation is dependant on the polarity and substrate temperature. Mg 
incorporation at higher Mg flux and higher temperature was found to be approximately 
twenty to thirty times less in N-polar GaN while compared to Ga-polar GaN. It was also 
shown that the Be incorporation in GaN by RF plasma MBE is much better than Mg 
doping and is also significantly less dependant on growth conditions.

Cheng et. al. [35] studied Mg–GaN growth by MBE on GaAs substrates. The growth 
rates of the samples were 0.3 \(\mu\text{m/hr}\). The dip in the Mg concentration close to the 
substrate epilayer interface was shown in the SIMS profile and was in close agreement to 
the work reported in Ref. [81]. In this model, it was assumed that the Mg incorporation 
was from a finite surface concentration and was concluded that high doping density was 
obtained initially by increasing the Mg flux but then it was saturated as the Mg surface 
concentration reached a finite value. Foxon et. al. [77] supported the incorporation of Mg 
from a surface concentration maintained during the growth by incident flux.
The effect of growth temperature on Mg doped GaN was reported in Ref. [34, 82]. Experiments reported in Ref. [34] were carried out in nitrogen rich condition with nitrogen flux of $\sim 8.3 \times 10^{14} \text{atoms/cm}^2 \cdot \text{s}$. Ga flux was maintained at $\sim 1.1 \times 10^{14} \text{atoms/cm}^2 \cdot \text{s}$, and the flux ratio were as follows: $J_{\text{Ga}} / J_{\text{Mg}} \sim 2$ and $J_{\text{Ga}} / J_{\text{N}} \sim 0.13$. SEM micrographs and AFM were used to study the surface morphology and the grain size as a function of substrate temperature. It was shown that at higher temperature, Mg doping promotes diffusion of Ga adatoms and enhances the layer-by-layer growth of GaN.

Myoung et. al. [82] compared the PL spectra of Mg-doped GaN films grown at two different temperatures. It was found that the films grown at high temperature had good crystalline quality and smooth surface while the films grown at low temperature had poor crystalline quality and rough surface.

The changes in the growth behavior of GaN due to the presence of Mg as a surfactant were studied by Mulo et. al. [83]. It was shown that the growth rate could be increased by 50% depending on the amount of Mg and III/V ratio. Daudin et. al. [84] found that Mg induces drastic changes in the surface diffusion and/or the residence time of both N and Ga. It was also found that in the nitrogen limited regime, Mg increases the amount of N available for the growth, due to the desorption rate of N atoms. In the Ga limited regime more Ga is available under fixed Ga flux value and the growth rate is increased.

Liliental-Weber et. al. [85] accomplished the doping of GaN by Si and Mg incorporation during the film growth. It was found that the post growth treatment is not required for the Mg-doped GaN films grown by MBE for electrically activating the dopant. Ng et. al. [86] observed the resistivity of Mg doped GaN films as a function of

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the Mg cell temperature. It was shown that at low temperatures, the incorporated Mg was not sufficient to compensate for the native defects. Increasing the Mg flux by four orders showed only an order of magnitude change in the hole carrier concentration of the epilayer. This behavior is explained based on inefficient incorporation of Mg due to the high vapor pressure of Mg [87]. The Mg concentration in the sample was measured using SIMS. The distribution of Mg in the epilayer was found to be homogeneous, but there was an accumulation of Mg close to the GaN and substrate interface. The accumulation of Mg was explained due to increased density of structural defects due to lattice mismatch at the interface.

Moustakas et. al. [46] worked on the growth and doping of GaN films and supported that p-type doping of GaN films grown by ECR assisted MBE can be obtained without any post growth annealing treatment. The hole carrier concentration at 300 K was found to be between $10^{18} - 10^{19} \text{cm}^{-3}$.

2.5 Models

The nature of the growth mechanism in lattice matched III-V compounds grown by MBE were identified for the first time by Madhukar et. al. [23]. Monte Carlo simulations were carried out for different Ga flux values and substrate temperatures, including additional pathways for arsenic dissociation reaction for GaAs MBE growth. It was shown that the basic nature of growth mechanism remains the same, but the quantitative behavior of specific consequences changes with the growth conditions. The quality of the interface for (100) growth of III-V semiconductor structures was controlled by the cation surface kinetics. Singh et. al. [24] studied the role of resonant Laser
enhanced surface kinetics in the MBE growth of compound semiconductors through Monte Carlo simulations.

A quasi-thermodynamic model was developed by Karpov et. al. [25] accounting for the kinetics of molecular nitrogen evaporation during the growth simulation of binary and ternary group III-nitrides. The growth processes for the binary nitrides were studied by analyzing the composition of the desorbed vapor species that were influencing the native defect formation in group III-nitrides. The specific features for this growth processes were analyzed. The approach was extended to study the growth of ternary nitrides. Two-drop behavior was found from the plot of growth rate of ternary nitrides and temperature.

A theoretical model [26] was proposed for the analysis of group III-nitrides, which accounts for a physisorption precursor of molecular nitrogen. The high thermal stability of nitrides were explained in relation with the desorption kinetics. The model was quasi thermo dynamic and the conditions of surface liquid formation of nitrides during growth were developed. A theoretical model to study the kinetics of surface processes during the GaN growth by MBE with ammonia was proposed by Karpov et. al. [27]. Desorption of Ga from the surface was considered as the first order reaction. Desorption of nitrogen involving in the association of nitrogen atoms was considered to be the second order reaction. The theoretical results were compared with the experimental results. The temperature dependence of the growth rate was shown to be of bell shaped.

Held et. al. [28] developed a rate equation model for GaN growth in the temperature regime where the surface morphology remains constant and the decomposition can be neglected. The adsorption of Ga on GaN (000\text{‾}) was studied by exposing the surface to incident Ga flux and the desorbing Ga flux versus time was measured. Desorption Mass
Spectroscopy was used to study the surface processes. It was shown that the growth could be modeled by the set of rate equations with some assumptions. Fu et. al. [36] proposed a rate equation approach for GaN growth in order to capture the essence of various experimental data in terms of physical model. All surface processes were assumed to be Arrhenius type. It was shown that the incorporation of Ga increases with increasing NH$_3$ overpressure, saturates at maximum Ga flux rate and attains a peak at 820°C.

Also a rate equation model for the GaN MBE growth using ECR nitrogen plasma was developed by Fu et. al. [29]. The model was similar to the one proposed in Ref. [36]. The unknown model parameters were found by fitting the results from simulation to the experimental values. It was found that as the ECR power was increased, the flux of the by-products were also increased which in turn resulted in the increase of Ga incorporation and saturated at a maximum rate. Sipe et. al. [30] modeled the GaN growth by MBE in the presence of low Mg flux using a set of rate equations. Ga limited and low – high nitrogen limited regimes were considered and it was assumed that Mg induces the incorporation of Ga. It was also assumed that the incorporation rate depends on the size of Ga cluster. It was shown that the exchange reaction between Ga and Mg aids in faster growth rate and the Ga cluster size dependence of growth rate results in non-monotonic behavior with Ga flux.

2.6 Devices

III-N devices have wide range of commercial applications including high-density optical data storage, full color displays, signal and automotive lighting, solar blind detectors, biological monitoring, high power electronics and underwater communication
In order to select a material for high power applications the characters that are to be noted are the power losses, switching losses in the high frequency and the critical breakdown field.

\( \text{GaN} \) exhibits Negative Differential resistance due to various effects that are reported in Ref. [3]. The device studies reported in Ref. [4, 5] have shown the possibility of realizing the NDR diodes. Pavlidis [3] has reviewed the transport characteristics of III-V nitrides and the technology of fabricating such two terminal NDR III-V diodes.

Paul et. al. [6] worked on several wide band gap compound semiconductors for superior high voltage unipolar power devices. It was shown that besides \( \text{SiC} \) and diamonds, the materials like \( \text{AlN, GaN} \) and \( \text{InN} \) and the intermetallics like \( \text{Ga_}\text{X} \text{In}_{1-\text{X}} \text{N} \) offers higher magnitude of on- resistance and potential for the operation of devices in higher temperatures. Khan et. al. [7] described the current/voltage characteristic collapse in the \( \text{AlGaN/GaN} \) heterostructure insulated gate field effect transistors, that were grown on sapphire substrates, during the high drain bias. They have shown that the devices exhibit both the low and high resistances before and after the application of high drain voltages.

Nichia chemical Industry introduced the high brightness blue LEDs based on \( \text{GaN} \) in 1994 [8]. The diodes are 100 times brighter than the \( \text{SiC} \) blue LEDs. Molnar et. al. [9] reported the growth of \( p-n \) homojunction LEDs using the Electron Cyclotron Resonance assisted Molecular Beam Epitaxy. The use of ECR technique is to reduce the hydrogen incorporation during the film growth. It was found that these devices emit light in the blue - violet spectrum and they do not require post-annealing treatment for efficient operation in contrast to the similar devices grown by MOCVD.
*InGaN* can be used in fabricating blue, green, amber, violet and red LEDs. Impurity doping in *InGaN* was performed in order to obtain blue emission spectra. By increasing the indium mole fraction in the active layer, LEDs of 2 cd brightness can be obtained. *InGaN/AlGaN* blue, green LED traffic lights are used in Japan, which consumes only twelve percent of the power when compared to the incandescent traffic lights. III-Nitride devices have a longer lifetime in the order of thousands of hours [10].

Red *InGaN* LEDs with the emission wavelength of 675 nm were fabricated by Nakamura *et. al.* [10]. In spite of large thread dislocations (TDs) introduced by GaN and sapphire mismatch, *InGaN* based LEDs have efficiency higher than that of conventional III-V compound semiconductors. Experiments were carried out to study the emission mechanism of GaN and *InGaN* quantum wells (QWs) by comparing their optical properties as a function of TD density. It was found that the net volume of the light emitting area is reduced by TDs. This negative impact is less effective in *InGaN* QWs where carriers are localized at certain potential. The localized states of an *InGaN* layer play a key role in the high efficiency of LEDs. When electrons and holes are injected into the active layer of LEDs, the carriers are captured by localized energy states before they are captured by the non-radiative recombination centers. Localized excitons with high binding energy are formed and they recombine radiatively.

Mukai *et. al.* [11] fabricated *InGaN* blue LEDs on Epitaxially Laterally Over Grown (ELOG) GaN substrates and observed blue shift with increasing forward current. *InGaN* single quantum well blue and green LEDs have a distinct mechanism of blue shift emission energy. Azuhata *et. al.* [12] and Takashi *et. al.* [64] investigated the mechanism that leads to this phenomena and concluded that it is due to the existence of tail energy.
states in band edges of InGaN.

Nakamura et. al. [13] was the first to develop the blue InGaN/AlGaN double heterostructures LEDs. Heterosturcture blue, green, amber and violet LEDs were fabricated by many researchers [14, 15, 16, 18, 13]. GaN homojunction $p-i-n$ photodiode was compared with AlGaN/GaN hetero junction $p-i-n$ detector and was found that the peaked response exhibited by homo junction near the band edge was absent in the hetero junction [19]. The degradation of blue AlGaN/InGaN/GaN LEDs subjected to the high electrical stress was studied by Daniel et. al. [17]. It was shown that GaN based LEDs are susceptible to electro migration failure under high current pulses.

The fabrication of GaN based blue laser diodes have made a revolutionary effect in the production of extremely high-density optical storage systems. GaN injection laser was first fabricated by Nakamura et. al. [63]. The device consisted of a $p-i-n$ junction with multiple quantum wells and with InGaN in $i$–region. This transistor comprises of a narrower bandgap base that is made of p-type 6H SiC. The normal Si transistor needs cooling fins to dissipate the heat generated while operating in high power. But the GaN/SiC HBT can operate at elevated temperatures without any cooling means. These devices have excellent inherent internal dissipation of heat.

GaN doped with erbium and oxygen is used for an electrically pumped 1.54$\mu$m laser in optical fiber communication [63]. They are very highly efficient than the narrower bandgap semiconductors. Hot electrons induced luminescence is found to be very efficient at room temperature in GaN doped with Er, O. Nakamura et. al. [20] fabricated InGaN multi quantum well structure Laser diodes. It was shown that the cleaved mirror facet showed a high output power under room temperature. The lifetime of the LDs at
constant output power of 5mW was found to be 160 hr. under CW operation at 50 °C ambient temperature.
3.1 Surface Processes

The MBE growth simulation of InGaN on wurtzitic GaN substrate oriented along [0001] and Mg doping of wurtzitic GaN were considered. The dynamic processes occurring on a surface riding physisorbed material layer (PM) and the surface of the crystalline epilayer play a crucial role in the growth and composition of compound semiconductors. The atoms or molecules of this layer are physisorbed on to the surface by Vander Waals type of binding, i.e., weakly bonded to the surface.

Several dynamic processes such as the adsorption of atom onto the crystal, the evaporation of atom out of it and the segregation of atoms from the bulk into the PM layer are considered for the PM layer. These processes are assumed to be Arrhenius type and are given by:

$$\tau_i = \frac{E_i}{kT} \tau_{o,i,e}$$

where $\tau_{o,i,e}$ represents the time constant for the process $i$, $E_i$ is the activation energy, $k$ is the Boltzmann constant and $T$ is the temperature in Kelvin.

The surface dynamic processes included for the epilayer are adsorption, evaporation, interlayer migration, intralayer migration and segregation. A schematic picture of these processes is shown in Figure 1 for InGaN and Figure 2 for Mg-GaN. Arrhenius type rate
equations are assumed for the rate of evaporation and migration of atoms, and are given by:

\[ R = R_0 e^{-\frac{E_{act}}{kT}} \]  

(2)

where \( R_0 \) is the frequency prefactor, \( E_{act} \) is the activation energy, \( k \) is the Boltzmann constant and \( T \) is the temperature in Kelvin.

3.2 Time Evolution Equations

3.2.1 InGaN Growth

The time evolution of the growing epilayer is described through the change of macro variables such as coverages of individual species resulting from the surface processes. The macro variables of growth are normalized with respect to the maximum number of possible atoms in the layer, and therefore have values between 0 and 1. Coverage is 1 when the layer is full and 0 when the layer is empty.

The layer coverages of \( Ga, N, \) and \( In \) in their respective layers are the macro variables describing the growth and are denoted as:

\[ C_{Ga}(2n) \]: layer coverage of \( Ga \) in the \( 2n^{th} \) layer,

\[ C_N(2n+1) \]: layer coverage of \( N \) in the \( 2n+1^{th} \) layer,

\[ C_{In}(2n) \]: layer coverage of \( In \) in the \( 2n^{th} \) layer,

\[ C_{Ga}^{PM}, C_N^{PM} \text{ and } C_{In}^{PM} \]: PM layer coverage of \( Ga, N \) and \( In \) respectively.  

(3)

where \( n \) is the layer index, with the \( Ga \) and \( In \) belonging to even numbered layers, and the \( N \) belonging to the odd numbered layers. Note that the coverages can have values
between 0 and 1, depending on how full a layer is.

The time evolution of the layer coverage of In in the 2\(n\)\(^{th}\) layer due to the various surface processes is given by:

\[
\frac{dC_{ln}(2n)}{dt} = \left[ C_N(2n-1) - C(2n) \right] \times \left[ J_{ln} + \frac{C_{ln,\text{phys}}}{\tau_{ln}} \right]
\]

(A1)

\[ + \left[ C_N(2n-1) - C(2n) \right] \times R_0 e^{\frac{E_{l,n}(2n+2)}{kT}} \left( \frac{C_{ln}(2n+2)}{C(2n+2)} \right) \]

\[ \times \left[ C(2n+2) - C_N(2n+3) \right]\]

(B1)

\[ + R_0 e^{\frac{E_{l,n}(2n-2)}{kT}} \left( \frac{C_{ln}(2n-2)}{C(2n-2)} \right) \times \left[ C(2n-2) - C_N(2n-1) \right] \]

\[ \times \frac{C_N(2n+1)}{C(2n+1)} \]

(C1)

\[ - R_0 e^{\frac{E_{l,n}(2n)}{kT}} \left( \frac{C_{ln}(2n)}{C(2n)} \right) \times \left[ C(2n) - C_N(2n+1) \right] \]

(D1)

\[ - R_0 e^{\frac{E_{l,n}(2n)}{kT}} \left( \frac{C_{ln}(2n)}{C(2n)} \right) \times \left[ C(2n) - C_N(2n+1) \right] \]

(E1)

where the term A1 represents the increase in \(C_{ln}(2n)\), resulting from adsorption of In from the incoming flux. The rate of adsorption is the product of the available sites for In incorporation on the surface, \([C_N(2n-1) - C(2n)]\), and the fluxes of In, \(J_{ln}\) from the molecular beam and \(\frac{C_{ln,\text{phys}}S}{\tau_{ln}}\) from the PM layer. The sticking coefficient of In is taken

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as unity. The term B1 describes the increase in $C_{In}^{2n}$ resulting from migration of In into the $2n^{th}$ layer from all the adjacent layers, i.e., $(2n + 2)$ and $(2n - 2)$. 

$[C(2n - 2) - C_N^2(2n - 1)]$ is the fraction of available sites for In in the $2n^{th}$ layer. In and Ga are the two possible elements in the cation sub-lattice. The total coverage of a cation layer, $C(2n + 2)$, is given by:

$$C(2n + 2) = C_{Ga}(2n + 2) + C_{In}(2n + 2)$$ \hspace{1cm} (5)

The fraction $\frac{C_{In}^{2n + 2}}{C(2n + 2)}$ is used to include only the In portion of all the cations in the layer for migration. The activation energy for In diffusion in the 2n-2th layer depends on the coverage of the layer and is given as:

$$E_{d, In}^{2n - 2} = E_{d, In, iso} + 6E_{In, In}C_{In}^{2n - 2} + 6E_{Ga, In}C_{Ga}^{2n - 2}$$ \hspace{1cm} (6)

where, $E_{d, In}$ is the activation energy of isolated atoms, and the second neighbor atom-atom pair interaction energies, $E_{Ga, In}$ and $E_{In, In}$, with a factor of six for the maximum possible number of neighboring atoms. Thus, when the coverage is very small, $E_{d, In}^{2n - 2}$ equals $E_{d, In, iso}$. $[C_N^{2n} - C(2n + 1)]$ is the portion of the $n^{th}$ layer available for diffusion of Ga/In from the neighboring layers. Term C1 represents the migration process opposite to that represented by the term B1. The terms D1 and E1 in Equation (4) describes the decrease in $C_{In}^{2n}$ resulting from the evaporation and segregation of In atoms, respectively, from the $2n^{th}$ layer. $\frac{C_{In}^{2n}(2n)}{C(2n)}[C(2n) - C_N^{2n} (2n + 1)]$ represents the fraction of In in the $2n^{th}$ exposed layer to the vacuum so that it is available for segregation or evaporation.
The descriptions of the activation energy for evaporation and segregation are similar to that of diffusion given by equation (4), except for the value for the isolated adatom. The equations are

\[ E_{c,ln}(2n) = E_{c,ln,iso} + 6E_{ln,ln}C_{ln}(2n) + 6E_{Ga,ln}C_{Ga}(2n) \]  

(7)

and

\[ E_{x,ln}(2n) = E_{x,ln,iso} + 6E_{ln,ln}C_{ln}(2n) + 6E_{Ga,ln}C_{Ga}(2n) \]  

(8)

for evaporation and segregation, respectively.

The time evolution of the layer coverage of \( In \) in the PM layer, \( \frac{dC_{ln}}{dt} \), is given by:

\[
\frac{dC_{i,phy}}{dt} = \left[ \left( J_i(1-S_i) \right) - \frac{C_{i,phy}}{\tau_{ev}^i} - \frac{C_{i,phy}S_i}{\tau_{ln}^i} + R_0e^{-\frac{E_i(2n)}{kT}} \frac{C_{i,phy}(2n)}{C(2n)} \right] \times [C(2n) - C_n(2n+1)]
\]  

(9)

where \( i \) represents \( In \) in this case and \( S_i \) is the sum of all crystal sites that are available for the incorporation of species in the appropriate sub-lattice. \( J_i \) is the molecular flux of the \( i^{th} \) species coming onto the substrate. The first term denotes the increase in PM coverage due to arrival of \( i^{th} \) species flux. The second and third term denotes the net loss of PM layer coverage due to evaporation and chemisorption. The last term denotes the gain in the PM layer coverage due to segregation.

A similar equation is written for physisorbed Ga and \( N \) without the segregation term and the \( N \) getting incorporated in the anion sub-lattice. As only monolayer coverage of the PM layer is effective in the surface dynamics, the sum of the coverage of Ga, \( In \) and \( N \) in the PM layer cannot exceed 1. \( R_0 \) is the frequency factor and \( E_i(2n) \) is the activation energy for the segregation of the \( i^{th} \) species from the crystal surface to the PM layer.
Equations similar to 4-9 are written for \( N \) and \( Ga \), except that the term \( E \), which represents segregation, is excluded from them.

The time evolution of the layer coverage of \( Ga \) in the 2n\(^{th} \) layer due to the surface processes is given by

\[
\frac{dC_{Ga}(2n)}{dt} = \left[ C_{Ga}(2n-1) - C(2n) \right] \times \left[ J_{Ga} + \frac{C_{Ga,phy}}{\tau_{m}} \right] \tag{A2}
\]

\[
+ \left[ C_{Ga}(2n-1) - C(2n) \right] \times R_{o}e^{\frac{E_{Ga,Ga}(2n-2)}{kT}} \left( \frac{C_{Ga}(2n+2)}{C(2n+2)} \right)
\times \left[ C(2n+2) - C_{Ga}(2n+3) \right] \tag{B2}
\]

\[
+ R_{o}e^{\frac{E_{Ga,Ga}(2n-2)}{kT}} \left( \frac{C_{Ga}(2n-2)}{C(2n-2)} \right) \times \left[ C(2n-2) - C_{Ga}(2n-1) \right] \tag{B2}
\]

\[
- R_{o}e^{\frac{E_{Ga,Ga}(2n)}{kT}} \left( \frac{C_{Ga}(2n)}{C(2n)} \right) \times \left[ C(2n) - C_{Ga}(2n+1) \right] \times \left[ C_{Ga}(2n+1) - C(2n+2) \right] + \left[ C_{Ga}(2n-3) - C(2n-2) \right] \times [C(2n-2) - C_{Ga}(2n-1)] \tag{C2}
\]

\[
- R_{o}e^{\frac{E_{Ga,Ga}(2n)}{kT}} \left( \frac{C_{Ga}(2n)}{C(2n)} \right) \times \left[ C(2n) - C_{Ga}(2n+1) \right] \tag{D2 (10)}
\]

The terms \( A2, B2, C2 \) and \( D2 \) are similar to \( A1, B1, C1 \) and \( D1 \) respectively.

The time evolution of layer coverage of \( N \) is given by:

\[
\frac{dC_{N}(2n+1)}{dt} = \left[ C(2n) - C(2n+1) \right] \times \left[ J_{N} + \frac{C_{N,phy}}{\tau_{m}} \right] \tag{A3}
\]

\[
+ \left[ C(2n) - C(2n+1) \right] \times R_{o}e^{\frac{E_{N,N}(2n+1)}{kT}} \left( \frac{C_{N}(2n+3)}{C(2n+3)} \right)
\times \left[ C(2n+3) - C(2n+4) \right]
\]

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where the terms $A_3$, $B_3$, $C_3$ and $D_3$ are similar to $A_1$, $B_1$, $C_1$ and $D_1$ respectively.

### 3.2.2 Mg-GaN Growth

The model used to describe Mg-GaN growth is same as the one used for InGaN growth except for the equation representing the activation energy of Mg diffusion. All the equations in InGaN except equations (7) and (8) are employed in this model with the subscript $In$ changed to $Mg$.

The activation energy for Mg diffusion in the $2n$-$2^{th}$ layer depends on the coverage of the layer and is given as:

$$E_d,Mg,u (2n-2) = E_d,Mg,iso,u + 6E_{Mg,Mg} C_{Mg} (2n-2) + 6E_{Ga,Mg} C_{Ga} (2n-2)$$

and

$$E_d,Mg,d (2n-2) = E_d,Mg,iso,d + 6E_{Mg,Mg} C_{Mg} (2n-2) + 6E_{Ga,Mg} C_{Ga} (2n-2)$$

where, $E_{d,iso,u}$ is the activation energy of isolated atoms moving upwards and $E_{d,iso,d}$ is the activation energy for atoms moving downwards, and the second neighbor atom-atom pair interaction energies, $E_{Ga,Mg}$ and $E_{Mg,Mg}$, with a factor of six for the maximum possible number of neighboring atoms. Thus, when the coverage is very small,
$E_{d,Mg}(2n-2)$ equals $E_{d,Mg,In}$. \([C_N(2n) - C(2n+1)]\) is the portion of the $n^{th}$ layer available for diffusion of Ga/Mg from neighboring layers. The differentiation made here for Mg migrating up or down is not done for In migration as described in section 3.2.1.

3.3 Computational details and model parameters

Three first order nonlinear differential equations are required for the description of the evolution of each bilayer of InGaN/Mg-GaN with one equation describing the time evolution of each of the normalized macro-variables. Three additional equations are required for describing the time evolution of the PM layer. For this work, a total of 243 coupled nonlinear first order differential equations are required to solve the simultaneous growth of 80 bilayers and the PM layer.

Fourth order Runge-Kutta method was used to integrate the system of equations with time steps of less than $10^{-6}$ s to compute the values of each of the macro-variables as a function of time for a growth time of 40 s. Similarly the activation energies for In, Ga and N evaporation process from the PM layer, $E_{ev}^{In}$, $E_{ev}^{Ga}$ and $E_{ev}^{N}$ respectively for InGaN and $E_{ev}^{Mg}$, $E_{ev}^{Ga}$ and $E_{ev}^{N}$ for Mg-GaN are assumed to be linearly dependent on their own coverage in the physisorbed layer [Table 1].

In the case of InGaN, in order to make the model simple, the 2nd neighbor interaction energies are kept the same and also the 1st neighbor interaction energy between III-V atoms are kept identical. In Mg-GaN the first neighbor interaction energy between Ga-N and Mg-N are made equal. The first neighbor interaction energy between Mg-Ga is made smaller and is equal to the second neighbor interaction energies. The prefactor of time constants for incorporation and evaporation processes are obtained according to the
Arrhenius equation and related to the activation energies. The evaporation, segregation and diffusion processes in the surface of the epilayer are assumed to be thermally activated and are modeled with the frequency factor, \( R_0 \) and activation energy. \( R_0 \) is linearly dependent on the substrate temperature, and is given by:

\[
R_0 = 2.08 \times 10^{10} \times T
\]  

(13)

This is based on the phonon frequency obtained using the equipartition energy principle. The frequency prefactor of diffusion processes are assumed constants. The frequency prefactor for segregation is considered to be linearly dependent on the substrate temperature, and is given by:

\[
R_{0,s} = 1.743 \times 10^{10} \times T
\]  

(14)

The segregation process from the PM layer is allowed only for \( In \) in the \( InGaN \) and only for \( Mg \) in \( Mg-GaN \). It is noted that \( R_{0,s} \) is smaller than the \( R_0 \) of evaporation and diffusion. The frequency factors for the other processes are:

\[
R_{0,\text{Ga}} = 4.38 \times 10^7 / s
\]

\[
R_{0,N} = 4.38 \times 10^7 / s
\]

\[
R_{0,Mg} = 2.4 \times 10^8 / s.
\]

(17)

3.4 Conversion among various flux units

The flux rates given in \( \text{atoms/cm}^2 \text{s} \) are converted to \( \left( \frac{ML}{s} \right) \). The conversion is done by multiplying the flux in \( \text{atoms/cm}^2 \text{s} \), with the area/site of the wurzitic structure.

\[
J \left( \frac{ML}{s} \right) = J \left( \frac{\text{atoms}}{\text{cm}^2 \text{s}} \right) \times \frac{\sqrt{3}}{2} a^2,
\]  

(18)
where \( a \) is the lattice constant. For \( GaN \) \( a = 3.189 \text{Å} \) and hence,

\[
J \left( \frac{ML}{s} \right) = 8.8072 \times 10^{-16} \times J \left( \frac{\text{atoms}}{cm^2s} \right)
\]  

(19)

For the wurzitic structure \( c = 5.185 \text{Å} \) and every monolayer is equal to \( \frac{c}{2} \) and hence,

\[
J \left( \frac{ML}{s} \right) = \frac{J \left( \frac{\text{nm}}{\text{min}} \right)}{\frac{c}{2} \left( \frac{\text{nm}}{\text{min}} \right)}
\]  

(20)
CHAPTER 4

RESULTS AND DISCUSSIONS

4.1 InGaN

The growth conditions for the simulations were obtained from Averbeck et al. [32]. The simulations were performed for two different nitrogen fluxes - 4.7 nm/min and 12 nm/min and for the various temperatures in the range of 500-700 °C. The In content versus temperature plot shown in Figure 3 is obtained for the $J_{Ga}/J_{N}$ ratio of 0.5 for various $J_{In}/J_{Ga}$ ratios in the range of 0.33-3.0. The behavior of the plots is within 1% agreement with the plots in Figure 1.a of Ref. [32]. It is found that for a given set of fluxes, below 580 °C, the fraction of In incorporated into the layer closely follows the ratio of In flux to the total flux, i.e., $J_{In}/J_{total}$, and the percentage of Indium content in the layer is constant. Above 580 °C, the segregation phenomenon begins to dominate the kinetics due to increased thermal energy and hence, In content decreases with increasing temperature. Above 640 °C, only a trace of In is found in the growing layer. The active segregation energy of In is found from our simulation to be 2.9 eV, which is in close agreement with the segregation energy given in Ref. [32].

The InN loss as a function of temperature for various flux conditions is shown in Figure 4. The InN loss rate here is computed as the difference between the In flux and the incorporation rate of In and hence, includes both the loss of In due to segregation and the
non-availability of N site. The simulations results show that for Group III rich conditions, the incorporation of Indium is limited by the nitrogen flux independent of the temperature. For N-rich growth conditions, at temperatures below 580 °C, the InN loss rate is insignificant as there is negligible segregation of In atoms due to lack of thermal energy necessary for overcoming the segregation barrier. Above 640 °C, the loss rate attains a maximum value of the In flux, $J_{In}$. The results of Figure 3 and Figure 4 are consistent.

Plots of percentage of Ga content in the layer as a function of temperature for various fluxes are shown in Figure 5. At lower temperatures the incorporation of Ga governed by the ratio of $J_{Ga}/J_{total}$. At temperature above 640 °C the Ga content is 100%, as all In segregates to the surface. Thus, the results of Figure 3 and Figure 4 are in consistent with that of Figure 5.

Plots of growth rate versus temperature are shown in Figure 6 for various flux conditions. For $J_N = 4.7 \text{ nm/min}$ and $J_{In}/J_{Ga} = 3.2$, $J_{In} + J_{Ga} = 9.9 \text{ nm/min}$, the growth rate was observed to be 3.2 nm/min at low temperatures and drops down to 1.6 nm/min at high temperatures. For $J_N = 4.7 \text{ nm/min}$ and $J_{In}/J_{Ga} = 1.1$, $J_{In} + J_{Ga} = 4.9 \text{ nm/min}$, the growth rate is 2.5 nm/min at low temperatures and decreases to 1.6nm/min. for high temperatures. At low temperatures, In contributes to the growth rate in accordance to the flux ratio of In to Ga. At high temperatures, In segregates and hence the growth rate stays constant, irrespective of the ratio of In and Ga flux.
4.2 Mg-GaN

The initial growth conditions are obtained from Myoung et al. [34]. The flux of nitrogen and flux ratios as given in [34] were $8.3 \times 10^{14} \text{atoms/cm}^2\text{s}$, $\frac{J_{\text{Ga}}}{J_{\text{N}}} \sim 0.13$ and $\frac{J_{\text{Ga}}}{J_{\text{Mg}}} \sim 2$ respectively. So the simulations are run for different temperatures in the range of 600-700 °C and at a particular flux rates of $\text{Mg}$, $\text{Ga}$ and $\text{N}$, such as $5.5 \times 10^{13} \text{atoms/cm}^2\text{s}$, $1.1 \times 10^{14} \text{atoms/cm}^2\text{s}$ and $8.3 \times 10^{14} \text{atoms/cm}^2\text{s}$ respectively.

Plot of Mg dopant concentration vs. layer number at 600 °C for various growth times is shown in Figure 7. For this plot, only layers that are full (i.e., $C_{\text{Mg}} + C_{\text{Ga}} = 1$) after a particular time are considered. The temperature was kept constant at 600 °C. Plots were obtained for various time periods of growth in seconds. The peak value in each plot denotes the maximum surface concentration at that particular layer at a particular time. The values to the left of the peak point represent the bulk concentration and to the right represent the surface concentration. Initially Mg concentration at the surface close to the substrate is high and as the epilayers grow the surface concentration of the growing epilayer increases. Mg concentration at the surface close to the substrate decreases, which shows that there is a segregation of Mg from layer to layer.

Plot of Mg concentration vs. layer number at 680 °C for various growth times is shown in Figure 8. Comparing Figures 7 and 8, it is observed that as the temperature increases, more and more Mg segregates to the surface and hence the bulk Mg concentration decrease with increase in temperature. This is in qualitative agreement with the experimental results in Ref. [34].
Due to computational limitations, it is not possible to simulate growth of films of thickness $\frac{1}{2} \mu m$. Thus an extrapolated scheme was adopted which is described below.

A cubic curve fitting is obtained for a plot of particular temperature in order to obtain the maximum $Mg$ concentration at various time periods of growth. Using the maximum Mg concentration obtained for various times, the constants a, b and c for the extrapolated equation

$$y_{\text{max}}(t) = a(1 - e^{-bt}) + c$$  \hspace{1cm} (21)

were obtained. Using equation 21, $y_{\text{max}}(t)$ values were obtained for $t = 5$ minutes. The constants for the extrapolated maximum $Mg$ surface concentration at various temperatures are listed in Table 2.

The extrapolated plot of maximum $Mg$ surface concentration vs. time is shown in Figure 9. The surface concentration increases with time and reaches a constant value, which implies that $Mg$ segregates to the surface and saturates at a particular concentration. The result is in close agreement with Figure 1.a in Ref. [35]. The extrapolated $Mg$ surface concentration vs. thickness of the crystal from the substrate, for various growth temperatures, is shown in Figure 10. The surface concentration increases from the substrate and reaches a maximum at the surface of the crystal. Also the saturated $Mg$ concentration increases with temperature and attains a peak value at $680 ^\circ C$. Above $680 ^\circ C$, the saturation value starts to decrease.

A plot of minimum and maximum $Mg$ concentration vs. temperature is shown in Figure 11. The minimum concentration implies the bulk $Mg$ concentration and the maximum concentration implies the surface concentration. The bulk $Mg$ concentration is obtained from the plots of dopant concentration vs. layer number for various
temperatures as shown in Figure 7 and 8. The point at which the plots start for various time periods is taken as the minimum bulk $Mg$ concentration. At low temperatures the thermally activated segregation energy that aids in layer-to-layer segregation is not dominant. But as the temperature increases, segregation also increases and the surface concentration of $Mg$ is maximum at 680 °C. Figure 11 is consistent with Figure 10.

When the temperature is raised above 750 °C there is a dip in the $Mg$ concentration near the surface layer as shown in Figure 11. This region is called as dopant depleted zone. The rate of these migration from one layer to the other depends on the availability of $Mg$ in the layers that are exposed to the vapor and the available sites in the layer to which the atoms migrate. For example, considering the $n^{th}$ layer, the ratio of atoms migrating to the $n^{th}$ layer to the atoms moving out of this layer is small. i.e. the rate of migration from the subsurface to the surface layer is larger than the rate of migration of atoms to the surface from the bottom layer. This gives rise to the deficiency of $Mg$ atoms near the surface layer. The dip shown in Figure 12 is in good qualitative agreement with the plots in Ref. [33, 34, 82].
CHAPTER 5

CONCLUSIONS AND RECOMMENDATIONS

The growth of InGaN and Mg-GaN by Molecular Beam Epitaxy is investigated with the rate equation model. The dependences of growth kinetics and In/Mg content on the temperature and flux conditions are analyzed using the simulation of the model. In the InGaN growth it is observed that the segregation phenomenon plays a major role in the surface kinetics and hence the In content of the grown layers. In segregation is found to be negligible below 580 °C. Above 640 °C, the segregation dominates the kinetics. This temperature dependence is found to be independent of the fluxes. In Mg-GaN growth, the phenomenon of segregation is justified with the simulation results and is in excellent agreement with the experimental results. The segregation of Mg is found to be dependant on the growth temperature. A dopant depleted zone near the surface is found to be formed during the growth at higher temperature. The observation is in qualitative agreement with the experimental data.

The following issues are recommended for further investigations:

- The dependence of Mg incorporation on the incident fluxes can be studied by simulating the model under various incident flux values.
- Theoretical models can be formulated for many experimental models that describe the incorporation behavior of Mg in Mg-GaN.
Table 1. Parameters, their symbols and values used in the simulation. X represents In in InGaN and Mg in Mg-GaN

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Parameter Name</th>
<th>InGaN (eV)</th>
<th>Mg-GaN (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_{XX}$</td>
<td>2nd neighbor atom-atom pair interaction energy for X cations.</td>
<td>0.08</td>
<td>0.08</td>
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<tr>
<td>$E_{Gd-Gd}$</td>
<td>2nd neighbor atom-atom pair interaction energy for Ga</td>
<td>0.08</td>
<td>2.00</td>
</tr>
<tr>
<td>$E_{NN}$</td>
<td>2nd neighbor atom-atom pair interaction energy for N</td>
<td>0.08</td>
<td>0.08</td>
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<tr>
<td>$E_{Gu-X}$</td>
<td>1st neighbor atom-atom pair interaction energy for Ga-X</td>
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<td>0.08</td>
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<tr>
<td>$E_{Ga-N}$</td>
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<td>2.0</td>
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<tr>
<td>$E_{X-N}$</td>
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<td>2.0</td>
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<tr>
<td>$E_{d,iso,Ga}$</td>
<td>Activation energy for diffusion for isolated Ga atom</td>
<td>1.2</td>
<td>1.2</td>
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<tr>
<td>$E_{d,iso,N}$</td>
<td>Activation energy for diffusion for isolated N atom</td>
<td>1.2</td>
<td>1.2</td>
</tr>
<tr>
<td>$E_{d,iso,X}$</td>
<td>Activation energy for diffusion for isolated X cation atom</td>
<td>1.2</td>
<td>$E_{d,iso,u} = 0.25$ $E_{d,iso,d} = 1.2$</td>
</tr>
<tr>
<td>$E_{e,iso,Ga}$</td>
<td>Activation energy for evaporation for isolated Ga atom</td>
<td>6.24</td>
<td>12.0</td>
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<tr>
<td>$E_{e,iso,N}$</td>
<td>Activation energy for evaporation for isolated N atom</td>
<td>6.24</td>
<td>6.24</td>
</tr>
<tr>
<td>$E_{e,iso,X}$</td>
<td>Activation energy for evaporation for isolated X cation atom</td>
<td>6.24</td>
<td>6.24</td>
</tr>
<tr>
<td>$E_{ev,Ga}$</td>
<td>Activation energy for the physisorbed Ga evaporation</td>
<td>$0.18 + 0.06C_{Ga,phy}$</td>
<td>$0.18 + 0.06C_{Ga,phy}$</td>
</tr>
<tr>
<td>$E_{ev,N}$</td>
<td>Activation energy for the physisorbed N evaporation</td>
<td>$0.18 + 0.06C_{N,phy}$</td>
<td>$0.18 + 0.06C_{N,phy}$</td>
</tr>
<tr>
<td>$E_{ev,X}$</td>
<td>Activation energy for the physisorbed X cation evaporation</td>
<td>$0.18 + 0.06C_{X,phy}$</td>
<td>$0.18 + 0.06C_{X,phy}$</td>
</tr>
<tr>
<td>$E_{in,Ga}$</td>
<td>Activation energy for the Ga incorporation</td>
<td>0.0</td>
<td>0.0</td>
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<td>$E_{in,N}$</td>
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<td>2.9</td>
</tr>
<tr>
<td>$E_{in,X}$</td>
<td>Activation energy for X cation incorporation</td>
<td>$0.5C_{X,phy}$</td>
<td>$0.5C_{X,phy}$</td>
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<td>$E_{seg,X}$</td>
<td>Activation energy for X cation segregation</td>
<td>2.9</td>
<td>2.9</td>
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Table 2 Constants for the extrapolated maximum $M_g$ concentration $y_{\text{max}}(t)$ at various temperatures

<table>
<thead>
<tr>
<th>Temperature ($^\circ$C)</th>
<th>$a$</th>
<th>$b$</th>
<th>$c$</th>
</tr>
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<tr>
<td>600</td>
<td>-2.2e-04</td>
<td>4.6e-04</td>
<td>0.0336</td>
</tr>
<tr>
<td>625</td>
<td>-8.9e-05</td>
<td>4.8e-04</td>
<td>0.0223</td>
</tr>
<tr>
<td>650</td>
<td>-1.5e-03</td>
<td>2.0e-03</td>
<td>0.0511</td>
</tr>
<tr>
<td>675</td>
<td>-2.6e-04</td>
<td>8.4e-04</td>
<td>0.0288</td>
</tr>
<tr>
<td>680</td>
<td>-3.1e-04</td>
<td>9.0e-04</td>
<td>0.0288</td>
</tr>
<tr>
<td>700</td>
<td>-1.2e-03</td>
<td>1.7e-03</td>
<td>0.0511</td>
</tr>
<tr>
<td>725</td>
<td>9.3e-05</td>
<td>2.3e-04</td>
<td>0.0105</td>
</tr>
</tbody>
</table>
FIGURE 1: A schematic picture of the surface processes in MBE growth of *InGaN*
FIGURE 2: A schematic picture of the surface processes in MBE growth of Mg-GaN.
FIGURE 3: Plots of Indium content in percentage vs the substrate temperature for the nitrogen fluxes of 4.7 nm/min and 12 nm/min, and various ratios of In and Ga fluxes.
FIGURE 4: Plots of InN loss rate vs temperature for the nitrogen fluxes of 4.7 nm/min and 12 nm/min, and various ratios of In and Ga fluxes.
FIGURE 5: Plots of Ga content in percentage vs substrate temperature for the nitrogen fluxes of 4.7 nm/min and 12 nm/min, and various ratios of In and Ga fluxes.
FIGURE 6: Plots of growth rate vs substrate temperature for the nitrogen fluxes of 4.7 nm/min and 12 nm/min, and various ratios of In and Ga fluxes.
FIGURE 7: Plot of dopant Mg concentration as a function of number of full layers at 600 °C for various growth times.
FIGURE 8: Plot of $Mg$ concentration as a function of number of full layers at 680 °C for various growth times.
FIGURE 9: The extrapolated data of the Mg surface concentration for increased duration of growth
FIGURE 10: The extrapolated data of Mg concentration as a function of layer thickness
FIGURE 11: Plot of bulk and extrapolated surface concentration of Mg for various growth temperatures.
FIGURE 12: Mg concentration vs. layer number at 750 °C. Note the dopant depleted zone for longer growth times.
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VITA

Graduate College
University of Nevada, Las Vegas

Irena Vidhya Mabel Stanley

Local Address:
4248 Chatham Circle
Las Vegas, NV 89119

Degree:
Bachelor of Science
Electronics and Communication Engineering, 2001
University of Madras, India

Thesis Title:
Theoretical Study of Segregation Kinetics of In in InGaN and Mg in Mg-GaN Grown by Molecular Beam Epitaxy

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Thesis Examination Committee:
Chairperson, Dr. Rama Venkat, Ph. D.
Committee Member, Dr. Shahram Latifi, Ph. D., P. E.
Committee Member, Dr. Sahjendra Singh, Ph. D.
Graduate faculty Representative, Dr. Ajoy K. Datta, Ph. D.