CHAPTER 4

SILICON ION IRRADIATION EFFECTS ON THE AlGaN/GaN HETEROSTRUCTURES

4.1 INTRODUCTION

Aluminum nitride (AlN), gallium nitride (GaN) and their ternary alloy aluminum gallium nitride (AlGaN) have emerged as important semiconductor materials for realizing applications in ultraviolet region of the electromagnetic spectrum as emitters, detectors, high-power and high frequency devices (Hirayama 2005). They are also well known for their hardness, high thermal conductivity, mechanical resistance to high temperature and aggressive chemical environment. These properties make them attractive for space applications (Mansouri et al 2008). In this chapter, swift heavy ions (SHI) irradiation at room temperature and low temperature (77 K) has been employed in order to study the radiation tolerance and defect resistance of AlGaN/GaN-heterostructures (HS).

Swift heavy ion beam effect on the materials depends upon the ion energy, fluence and ion species. The SHI interaction with material results in the modifications of structural, optical, electrical and surface morphological characteristics and applied properties (Jain et al 2011).

4.1.1 Interaction between Ions and Materials

Energetic ions deposit extremely high localized density of energy to the material, in a very short time ($\sim 10^{-17}$ to $10^{-15}$ s) within a very small
(\sim 10^{-17} \text{ to } 10^{-16} \text{ cm}^3) \) volume (Avasthi et al 2011). Depending on the energy density, the melting point of the materials can be exceeded in a region around the ion trajectory. During irradiation, SHI penetrates the material and interacts with electrons and the nuclei of the material. At high energy, the ions lose their energy predominantly by interacting with the electrons of the atoms constituting the material; at low energy, ions interacts with the nuclei of the atoms. This energy loses occur mainly due to two nearly independent processes:

(i) elastic collisions with the nuclei known as nuclear energy loss \( S_n \), which dominates at an energy of about 1 keV/amu

(ii) inelastic collisions of the highly charged projectile ion with the atomic electrons of the material known as electronic energy loss \( S_n \), which dominates at an energy of about 1 MeV/amu or more

In the inelastic collision, the energy is transferred from the projectile to the atoms through excitation and ionization of the surrounding electrons. The amount of electronic loss in each collision varies from tens of eV to a few keV per angstrom (Å).

The effect of ion implantation (Kucheyev et al 2001) and ion irradiation (Look 1997, Kucheyev et al 2004, Premchander et al 2006, Suresh Kumar et al 2006, Suresh et al 2011 and Varadarajan et al 2006) on GaN epitaxial layers have been extensively studied than other III-nitride materials. However, irradiation studies on AlGaN/GaN-heterostructures (HS) are very limited and it is imperative (Sonia et al 2006, 2008 and Howgate et al 2012).
4.2 EXPERIMENT

4.2.1 Pelletron

Ion irradiation facility has been availed at the Inter-University Accelerator Centre (IUAC), New Delhi. IUAC pelletron has a 15 UD tandem electrostatic accelerator, capable of accelerating ions with energies from 50 MeV to 200 MeV (Kabiraj 2007). The digit 15 stands for 15 MV terminal voltage and UD stands for Unit Double. The pelletron belongs to a class of accelerators known as tandem Van de Graff accelerator. A schematic diagram of the pelletron is shown in Figure 4.1.

The pelletron with a vertical geometry is installed in a stainless steel tank, which is 26.5 m long and 5.5 m in diameter. It is filled with sulfur hexa-fluoride insulating gas at a pressure of about 6-7 bar. In the middle of the tank, high voltage terminal of about 1.52 m in diameter and 3.81 m in height is situated and it can be charged to a potential from 4 to 16 MV. This terminal is connected to the tank vertically through ceramic titanium tubes known as the accelerating tubes. Potential gradient is maintained with the help of these tubes.

Negative ions are produced and pre-accelerated to about 250 keV by a sputter type ion source known as MC-SNICS (Multi-Cathode Source of Negative Ions by Cesium Sputtering). The ions of different masses are analyzed by a 90° dipole magnet called injector magnet and are turned in a vertically downward direction towards the terminal. On reaching the terminal, they pass through a stripper (C-foil or N₂ gas) which removes some electrons from the negative ions, thus changing them to positive ions. On further acceleration, they proceed towards the bottom of the tank at ground potential.
The final energy of the ions emerging from the accelerator is given by the equation below

\[ E = \{V_T (1+q)\} \text{MeV} \]  

(4.1)

where

\( V_T \) is the terminal potential in MV,

\( q \) is the charge state of the ion after stripping.
These high energy ions are analyzed in energy with the help of a 90° bending magnet known as analyzer magnet.

### 4.2.2 Beam Lines

IUAC has totally seven beam lines. Among them, one beam line is dedicated for material science. The ions are directed to the desired experimental beam line with the help of a multiport switching magnet which can deflect the beam to anyone of the seven beam lines in the beam hall. The whole beam line of the accelerator is in ultra high vacuum (UHV). The vacuum in the beam line is in the order of $10^{-9}$ torr. During the passage of ions through the accelerator beam line, the ion beam is kept centered and focused using steering magnets and quadrupole triplet magnets. The beam is visually monitored by the glow on the quartz and beam profile monitors (BPM). The beam current is measured by means of Faraday cups. The material science beam line is at 15° to the right with respect to the zero degree beam line. Material science beam line has three chambers and these are connected one after another as shown in Figure 4.2. In the high vacuum chamber, most of the irradiation and elastic recoil detection are carried out. The chamber is made up of stainless steel. The vacuum in the chamber is created by a turbo molecular pump. The chamber vacuum during the irradiation experiment is $6\times10^{-6}$ torr. The reason for the vacuum environment is to avoid any collision of the particle (beam) with gas molecules.

Figure 4.3 shows the irradiation chamber and the target ladder (where the samples have to be mounted). After mounting the sample, the ladder has to be inserted into the chamber for irradiation. There is a provision available in the top of the ladder to pour liquid nitrogen for performing low temperature irradiation experiments. A stepper motor in conjunction with suitable mechanical assembly is used to control the up and down motion of
the ladder. This up and down motion can also be done from the remote data acquisition room using an electronic control panel. The sample position can be aligned with respect to the ion beam by first looking at the luminescence of the beam on the quartz crystal.

![Schematic diagram of Materials Science beam line at IUAC](image)

**Figure 4.2 Schematic diagram of Materials Science beam line at IUAC**

Then, the sample is brought to the position of the quartz, by marking on the screen. The position of the quartz and samples are observed using close circuit television (CCTV) kept in data room. With the help of a magnetic quadrupole and a steerer, the beam is focused on the target. For irradiation, the beam is scanned in ‘x’ and ‘y’ direction over a maximum area.
of 10×10 mm² with the help of a magnetic scanner. The scanning ensures the uniformity of irradiation over the whole area of the sample.

Figure 4.3 Irradiation chamber and the sample ladder used for the irradiation of AlGaN/GaN heterostructure samples
A cylindrical enclosure of stainless steel (suppressor) surrounds the sample ladder, which is kept at a negative potential of 120 V. This enclosure suppresses the secondary electrons coming out of the sample during irradiation. An opening in the suppressor allows the ion beam to fall on the sample. The total number of the particles/charges falling on the sample can be estimated by a combination of the current integrator and the pulse counter, from which the irradiation fluence can be measured. The samples (1cm ×1cm) to be irradiated are mounted on the four sides of the target ladder (on copper block), which are separated from each other by a distance of about 15 mm. The time or counts can be calculated for the desired ion fluence for each sample using the following relation.

\[
\text{Time (T)} = F \times \frac{\text{Exposed Area (A)}}{I} \times 6.25 \times 10^9 \text{ sec} \quad (4.2)
\]

Equation 4.2 can also be written as \( T = [F \times A/I] \times e \)

where,

I is the beam current in (pnA)

\( e \) is the electron charge \((e = 1.6 \times 10^{-19})\)

A is the Area \((1 \text{ cm}^2)\)

(or)

\[
\text{Number of counts} = \text{Fluence} \times q \times A \times e / \text{scale} \quad (4.3)
\]

q is the charge state of ion

Scale – current scale in nA

In this study, AlGaN/GaN heterostructures (HS) were grown by Metal Organic Chemical Vapour Deposition (MOCVD) technique on 2 inch diameter sapphire (0001) substrates. Figure 4.4 shows the cross-sectional view of sample structure. The AlGaN/GaN growth was initiated with low temperature GaN nucleation layer on the sapphire substrate, followed by GaN
buffer layer with the thickness of 1.5 μm and 65 nm thick AlGaN layer. The growth temperature of GaN buffer and AlGaN layer have been maintained at 1040 °C and 1080 °C respectively. The grown sample was cut into pieces of dimensions about 10 mm × 10 mm. The pieces were mounted on the target ladder. A beam current of 1 pnA (particle nano ampere) has been maintained throughout the irradiation experiments.

120 MeV silicon (Si$^{9+}$) ion with the fluence of $5 \times 10^{12}$ ions/cm$^2$ has been used to irradiate the AlGaN/GaN-HS samples at room temperature (RT) and low temperature (LT) (liquid nitrogen temperature - 77 K). Pristine and irradiated AlGaN/GaN-HS samples were characterized using high resolution X-ray diffractometer (HRXRD), atomic force microscopy (AFM) Raman spectroscopy, and photoluminescence (PL).

4.3 RESULTS AND DISCUSSION

4.3.1 In-Situ Reflectance

Growth of AlGaN/GaN-HS has been monitored by the in-situ reflectance measurement using a laser source of 635 nm wavelength. Figure
4.5 reveals the reflectance peaks of GaN buffer and AlGaN layers as the function of time. The amplitude of GaN buffer layer reflectance peaks has been found constant after the recovery period (from the second oscillation of GaN buffer layer) to the end of growth. This pattern gestures the two dimensional growth of GaN with smooth surface. In addition to this, a clear reflectance peak was observed for AlGaN layer. It has been found to disclose abrupt interface between the GaN buffer and AlGaN layer. The small amplitude of AlGaN reflectance peak should be attributed to the low thickness of AlGaN layer.

GaN buffer layer thickness has been calculated as 1.5 microns from the reflectance peak. However, it has not been possible to estimate the thickness of AlGaN layer from the in-situ reflectance due to the low amplitude peak. The thickness and composition of AlGaN were estimated using HRXRD.

Figure 4.5  In-situ reflectance pattern of AlGaN/GaN-HS versus growth time
4.3.2 Stopping and Range of Ions in Matter (SRIM)

SRIM calculations have been carried out to understand the role of energy loss mechanisms, either by elastic collision (nuclear energy loss $S_n$) or by inelastic collision (electronic energy loss $S_e$) during the silicon ion interaction with AlGaN/GaN-HS samples.

Figure 4.6 SRIM plot shows electronic and nuclear energy loss with respect to the energy of silicon ion with ions projected range in the AlGaN/GaN-HS sample
Figure 4.6 depicts the SRIM plot of energy versus loss. The nuclear energy loss of 120 MeV Si$^{9+}$ ions is 3 orders of magnitude smaller than the electronic energy loss in AlGaN/GaN-HS. It is obvious to note that the maximum energy deposited to the AlGaN/GaN-HS has been mainly due to electronic energy loss during the passage of ions through the sample. The projected silicon ion range at 120 MeV energy is about 40 microns, greater than the thickness of the AlGaN and GaN layers. It denotes that AlGaN/GaN-HS suffers uniformly in the irradiation.

4.3.3 HRXRD Experimental Results

In order to study the structural quality of pristine and silicon ion irradiated AlGaN/GaN-HS samples, 2Theta scan, symmetric and asymmetric X-ray rocking curves (XRC), reciprocal space map (RSM) and omega-2Theta scan have been performed. Figure 4.7 shows the 2Theta plot of AlGaN/GaN-HS samples versus intensity.
Appearance of only (002) reflections of GaN and AlGaN indicates that there is no other phase formation. 2Theta position of GaN (002) peak has been obtained at 34.55 °, which is in agreement with the standard 2Theta value of (002) GaN layer peak (ICSD 50-0792). However, it has been worth noticing that GaN (002) peak positions are identical for pristine and irradiated samples. The reduction of the GaN peak intensities in the irradiated samples reveals the existence of irradiation induced damage. A small shift has been observed in AlGaN (002) peaks towards higher angle, from 34.9 (pristine) to 34.95 degrees for the irradiated samples. It is found to show the AlGaN layer in the heterostructure samples undergo radiation induced compressive stress.

Table 4.1 shows the X-ray rocking curve (XRC) full width half maximum (FWHM) values of (002) and (102) reflections for the GaN and AlGaN layers separately from the pristine and irradiated AlGaN/GaN-HS. Both the FWHM of (002) and (102) have found to be increased for the Irradiated AlGaN/GaN-HS samples.

<table>
<thead>
<tr>
<th>Ions</th>
<th>GaN (002) arcsec</th>
<th>GaN (102) arcsec</th>
<th>AlGaN (002) arcsec</th>
<th>AlGaN (102) arcsec</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pristine</td>
<td>363</td>
<td>718</td>
<td>332</td>
<td>733</td>
</tr>
<tr>
<td>120 MeV 28 Silicon(^{9+}) (RT)</td>
<td>379</td>
<td>837</td>
<td>352</td>
<td>786</td>
</tr>
<tr>
<td>120 MeV 28 Silicon(^{9+}) (77K)</td>
<td>376</td>
<td>873</td>
<td>362</td>
<td>833</td>
</tr>
</tbody>
</table>
The XRC results elucidate that the damage induced by the silicon ion irradiation is pronounced in the low temperature than the room temperature irradiated sample.

4.3.3.1 Reciprocal space map

X-ray reciprocal space map is frequently used to study the structural properties of epitaxial films. Reciprocal space map (RSM) is a two dimensional measurement that combines and records 2Theta and omega (\(\omega\)) scans simultaneously.

RSM can be performed around symmetrical and asymmetrical reflections of the sample. In addition to that, shape and positions of the reciprocal lattice points or the intensity contour plots reveal more information about mismatch, strain state, relaxation and mosaicity, etc. The horizontal and vertical positions of the lattice points exhibit information about the in-plane and out-of-plane lattice parameters, respectively. Also, it has been related to their lattice mismatch. In particular, the RSM can be used to determine whether the grown layers are fully strained or pseudomorphic, partially strained, or fully relaxed with respect to substrate. If the lattice points of the layer are vertically aligned with that of the substrate, they are found to correspond to a high quality pseudomorphic layer. For the partially strained or fully relaxed layers, the position of layer’s lattice points will not be in alignment with substrate lattice points. Figure 4.8 shows the asymmetric (114) plane reciprocal space map of pristine and irradiated AlGaN/GaN-HS samples. The asymmetric plane RSM explicit the strain state between the GaN layer and AlGaN layer. The (114) plane RSM of pristine and irradiated samples exhibit GaN and AlGaN peaks, which are vertically aligned.
Figure 4.8 Reciprocal space mapping of AlGaN/GaN-HS. (a) Pristine, (b) silicon ion irradiated at room temperature and (c) silicon ion irradiated at low temperature (77 K)

The parallel and perpendicular mismatch between the AlGaN and GaN layers present in the pristine and irradiated AlGaN/GaN-HS samples have been derived from the position of corresponding lattice points. The parallel mismatch \((Q_X^S - Q_X^L)/Q_X^L\) is found to be almost zero and the perpendicular mismatch \((Q_Y^L - Q_Y^S)/Q_Y^L\) is found to be around 1%. Where \(Q_X^S, Q_Y^S\) and \(Q_X^L, Q_Y^L\) are the ‘X’ and ‘Y’ co-ordinates of substrate (here GaN) and layer (AlGaN) respectively. It confirms that the AlGaN layers are fully strained on the GaN layers for the pristine and irradiated samples. However, slight variation in the lattice points contour of AlGaN layers and appearance of spots at the Figure 4.8 (b) and (c) explains the radiation induced structural defects.
Figure 4.9 Omega – 2Theta scan of AlGaN/GaN-HS. (a) pristine, (b) silicon ion irradiated at room temperature and (c) silicon ion irradiated at low temperature (77 K).

The composition of aluminum (Al) in the AlGaN layers has been estimated by the omega-2Theta scan. Figure 4.9 depicts the omega-2theta scan of pristine and irradiated AlGaN/GaN-HS samples. There is no change in
the composition of aluminum in the AlGaN layers after irradiation. The aluminum composition is estimated as 20% by fitting the experimental curve. It may be attributed to the chemical stability of the AlGaN layers upon irradiation.

However, the thickness of the AlGaN layers after irradiation has been found increased. The thickness of the AlGaN layer in the pristine AlGaN/GaN-HS is estimated as 65 nm, whereas the AlGaN layer thickness in the irradiated AlGaN/GaN-HS has been determined as 75 nm. It can also be clearly observed from the AlGaN layers peak width in the irradiated AlGaN/GaN-HS samples (see Figure 4.9 (b) and (c)).

4.3.4 Atomic Force Microscopy

Figure 4.10 shows the three dimensional AFM images of the pristine and irradiated AlGaN/GaN-HS samples. All scans are performed in 10 \( \mu \text{m} \times 10 \mu \text{m} \) area. The clear steps and terraces have been observed on the pristine AlGaN/GaN-HS samples showing the smooth morphology (Figure 4.10 a). After the silicon ion irradiation at RT, three dimensional island like clusters are found to be formed on the surface (Figure 4.10 b). On the other hand, clusters size has been noticed to be increased after the silicon ion irradiation at LT (Figure 4.10 c).

During the irradiation a huge amount of kinetic energy transferred to the material. It is found to increase the local lattice temperature above the melting point of the material. The temperature increase is then followed by a quenching \((10^{13} - 10^{14} \text{ K/s})\). In a few pico seconds, the material is found to melt and solidify again. During this process, clusters are found to be formed on the surface of the AlGaN layers in the range of few tens to hundreds of nanometer dimension. These clusters may also have been originated due to thermally induced agglomeration. The size of the clusters
formed at room temperature irradiated sample is found to vary from 25 to 90 nm. Whereas huge variation has been observed in the clusters size of low temperature irradiated sample in the range of 60 to 800 nm.

Figure 4.10 AFM images of AlGaN/GaN-HS. (a) pristine, (b) silicon ion irradiated at room temperature and (c) silicon ion irradiated at low temperature (77 K). All scans are 10 μm × 10 μm

The formation of big clusters is found to be predominant in LT-irradiated sample due to very rapid quenching. The root mean square (rms) surface roughness has been found as 0.5 nm, 1.7 nm and 5.6 nm for pristine AlGaN/GaN-HS, silicon ion irradiated AlGaN/GaN-HS at RT and at LT respectively. The 10 nm increase in the thickness of the AlGaN layer (estimated from omega-2Theta scan) might be understandable by the modifications of the AlGaN layer surface due to irradiation.

4.3.5 Raman Spectroscopy

Figure 4.11 shows the Raman spectra of pristine and silicon ion irradiated AlGaN/GaN-HS samples. Raman spectra have been recorded in the backscattering geometry. In this particular geometry, the allowed $E_2$ (high) and $A_1$ LO phonon modes has been found to appear for pristine and irradiated
samples. The $E_2$ (high) phonon mode is the most commonly used mode to monitor stress and strain in III–V/nitride semiconductor structures (Sarua et al 2002).

![Raman spectra of pristine and irradiated AlGaN/GaN-HS samples](image)

Figure 4.11 Raman spectra of pristine and irradiated AlGaN/GaN-HS samples

The position of $E_2$ (high) and $A_1$ (LO) phonon mode for unstrained GaN are 567 cm$^{-1}$ and 734 cm$^{-1}$ respectively. $E_2$ (high) phonon mode of GaN layer in the pristine and irradiated samples has been obtained at 570 cm$^{-1}$ (see Table 4.2). The shift of GaN $E_2$ (high) mode towards the higher frequency side has been related to the residual compressive stress and in turn the
pseudomorphic AlGaN layer on the GaN layer might also experience the same compressive stress. This result shows the sensitivity of Raman $E_2$ (high) phonon mode in exploring the stress and strain between the substrate and epilayers. Consequently, the absence of other/forbidden phonon modes [$A_1$ (TO) and $E_1$ (TO)] in the Raman spectra is found to ascertain that the clusters formed on the AlGaN layers has also been oriented along (002) direction. Table 4.2 discloses the positions of $E_2$ (high) and $A_1$ LO phonon modes of GaN and AlGaN layers present in the pristine and irradiated AlGaN/GaN-HS.

**Table 4.2 $E_2$ (high) and $A_1$ LO phonon modes of GaN and AlGaN**

<table>
<thead>
<tr>
<th>Sample</th>
<th>GaN $E_2$ (high) phonon mode (cm$^{-1}$)</th>
<th>AlGaN $E_2$ (high) phonon mode (cm$^{-1}$)</th>
<th>GaN $A_1$(LO) phonon mode (cm$^{-1}$)</th>
<th>AlGaN $A_1$(LO) phonon mode (cm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pristine</td>
<td>570</td>
<td>576</td>
<td>735</td>
<td>750</td>
</tr>
<tr>
<td>120 MeV 28 Silicon $^9+$ (RT)</td>
<td>570</td>
<td>577</td>
<td>734</td>
<td>749</td>
</tr>
<tr>
<td>120 MeV 28 Silicon $^9+$ (77K)</td>
<td>570</td>
<td>577</td>
<td>737</td>
<td>750</td>
</tr>
</tbody>
</table>

The $E_2$ (high) and $A_1$ (LO) phonon modes position of GaN and AlGaN layers in the pristine and irradiated AlGaN/GaN-HS samples are found almost constant. However the intensity of GaN and AlGaN phonon modes peaks has been found to reduce drastically in LT irradiated AlGaN/GaN-HS sample. It certainly confirms that the creations of defects are predominant at LT irradiation than RT irradiation due to rapid quenching.
4.3.6 Photoluminescence Studies

Figure 4.12 shows the photoluminescence spectra for the pristine and irradiated AlGaN/GaN-HS samples. Defect bands (DB) have been found to be dominant in the irradiated sample than the near band edge (NBE) emission of GaN and AlGaN layers.

![Photoluminescence spectra](image)

**Figure 4.12 Photoluminescence spectra of pristine and irradiated AlGaN/GaN–HS samples**

GaN NBE emission has been obtained at 3.42 eV for the pristine and irradiated samples without any shifts. Nevertheless, NBE emission of AlGaN layers present in the pristine, RT and LT irradiated AlGaN/GaN-HS is found as 3.86 eV, 3.88 eV and 3.93 eV respectively. With respect to the AlGaN NBE emission in the pristine sample, a shift towards higher energy
side of about 12 meV and 52 meV are observed for the AlGaN layers NBE emission in the RT and LT silicon ion irradiated samples.

The NBE emission shift towards higher energy side is noticed, when compressive strain acts along the in-plane direction. The compressive strain of AlGaN layers exhibited in the NBE emission for the irradiated samples has been found well in accordance with the XRD-2Theta scan results. The higher energy shift of AlGaN NBE emission can be attributed to the presence of strained clusters on the surface of AlGaN layers after irradiation. Table 4.3 discloses the ratio between the near band edge emission and defect band emissions of the GaN and AlGaN layers in the pristine and irradiated AlGaN/GaN-HS. It clearly depicts the optical quality of AlGaN layers and GaN layers after the irradiation has been drastically reduced.

**Table 4.3**  
Intensity ratio of NBE and Defect band emissions for GaN and AlGaN layers presents in the pristine and irradiated AlGaN/GaN-HS

<table>
<thead>
<tr>
<th>Ions</th>
<th>Ratio between GaN NBE and DB</th>
<th>Ratio Between AlGaN NBE and DB</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pristine</td>
<td>5</td>
<td>1.38</td>
</tr>
<tr>
<td>120 MeV  28 Silicon$^{9+}$ (RT)</td>
<td>0.16</td>
<td>0.22</td>
</tr>
<tr>
<td>120 MeV  28 Silicon$^{9+}$ (77K)</td>
<td>0.14</td>
<td>0.15</td>
</tr>
</tbody>
</table>
4.4 CONCLUSION

HRXRD studies have been noted to reveal that there are no compositional changes in the AlGaN layers and no additional phase formations in the AlGaN/GaN-HS after irradiation. FWHM of (102) reflection has been found to broaden for irradiated AlGaN/GaN-HS sample whereas the (002) FWHM values shows only slight increase. It depicts that the edge dislocations of AlGaN and GaN layers are increased in AlGaN/GaN-HS samples upon irradiation. AFM images depict the formation of nano clusters after irradiation. The size of the clusters is found to be increased at the low temperature irradiation and in turn rms surface roughness values are also found to increase. Predominant defect band emission in the PL spectra shows that the optical quality is highly deteriorated for the irradiated AlGaN/GaN-HS samples. Also the contribution of clusters in shifting the AlGaN NBE emission towards the higher energy is apparent. Raman spectroscopy depicts only the allowed phonon modes for the backscattering geometry. This indicates that the clusters observed in the surface of irradiated AlGaN/GaN-HS samples are only in (002) orientation. On the whole, it has been observed that the low temperature irradiation decreases the radiation tolerance and increases the defects and surface roughness of the AlGaN/GaN-HS than the room temperature irradiation.
CHAPTER 5

SUMMARY AND SUGGESTIONS FOR FUTURE WORK

5.1 SUMMARY

This thesis has been broadly classified into two parts. One part is dedicated to the growth of AlN epilayers using HVPE and MOCVD techniques. These AlN layers can serve as the template or the buffer layers for the AlGaN based deep ultraviolet (DUV) light emitting device structures. Another part focuses on studying the radiation tolerance and defect resistance of AlGaN/GaN-heterostructures (HS) upon silicon ion irradiation at room temperature and low temperature (77 K).

The potential effect of using low temperature NLs for the growth of AlN layers by high temperature HVPE has been examined. AlN nucleation layer deposition temperature has been varied as 650 °C, 750 °C and 850 °C. Initially, as-grown NLs were first characterized with X-ray reflectivity to know the thickness of NL. The density and size of nucleation islands have been studied using the atomic force microscopy (AFM). Then, the same samples have been reloaded into the reactor to perform high temperature (HT)-treatment. NLs were recrystallized at 1080 °C and further ramped up to 1200 °C called as HT- treatment. The HT-treated NLs have been again assessed by XRR and AFM to study the changes in the thickness and nucleation island’s density and size. It has been concluded that AlN-NL desorption at 1200 °C and rearrangement of the AlN-NL surface during high temperature treatment are existent.
Afterwards growth of HT-AlN layers with nucleation layers have been performed using HT-HVPE at 1200 and 1400 °C. The grown HT-AlN films with NLs deposited at different temperatures were characterized by X-ray diffraction (XRD) using θ-2θ scan, rocking curve (XRC) of the (002) peak, Raman and photoluminescence (PL) spectroscopies. Surface morphology was accessed by atomic force microscopy (AFM) and field emission scanning electron microscopy (FE-SEM). The thickness of all the HT-AlN samples was found to be approximately 2 micrometers. Improvement in structural quality, surface morphology and epitaxial growth along c-axis for the HT-AlN films has been observed when increasing the NLs deposition temperature from 650 to 850 °C. Finally, a comparison has been made between the quality of HT-AlN films grown at 1200 °C and 1400 °C by employing 850 °C NLs (optimal) process parameters. Two kinds of surfaces such as cracked surface and specular surface have been observed from the HT-AlN sample grown at 1400 °C. The difference between the thermal expansion coefficient of sapphire and AlN has been attributed to the formation of cracks. However, the quality of the HT-AlN layers has been found increased at 1400 °C.

Significance of initial growth stages such as pre-flow of trimethyl aluminum (TMAI) and ammonia (NH₃) prior to NL growth, direct deposition of NL (without pre-flows), influence of NL growth temperatures and the role of NIs coalescence on the surface morphology and structural quality of AlN layers grown by metal organic chemical vapor deposition (MOCVD) system have been systematically investigated. All the AlN samples have been grown with a V/III ratio of 512 and thickness of about 2 μm, after the recrystallization of nucleation layers. Initially, three AlN layers were grown on nucleation layers (NLs) deposited at 850 °C with a V/III ratio of 1024. Thickness of NLs was around 30 nm. Only the pre-flow conditions prior to the growth of NLs at 850 °C have been modified to optimize the initial
growth condition. Structural quality and surface morphology of these AlN layers were investigated by high resolution X-ray diffraction (HR-XRD) and atomic force microscopy (AFM) respectively. It has been found that direct deposition of AlN nucleation layer (without pre-flows) is the best condition for the growth of actual AlN layers at 1300 °C. After optimizing the initial growth condition, AlN layers were grown with only variation in the nucleation growth temperatures. Other growth parameters such as recrystallization temperature of AlN-NLs ($T_r$), growth temperature ($T_g$) of AlN layers, V/III ratios of AlN-NLs (1024) and AlN layers (512) were kept identical.

Increasing the NL growth temperature from 850 to 950 °C has been found to improve the quality of AlN layers. AlN islands diameter of about 80-100 nm has been formed at nucleation temperature of 950 °C. It is found to tend and coalesce uniformly to initiate the required two dimensional AlN layer growth in the subsequent process. Further increase in the AlN-NL growth temperatures from 950 to 1250 °C has been found to detriment the surface morphology and structural quality of AlN layers. In order to understand the effect of NLs deposition temperature, as grown NLs were examined by AFM. High variation in nucleation islands (NIs) size, height and insufficient coverage of NIs on the surface of sapphire has been noted to lead to uneven coalescence and it has been found to decrease the quality of AlN layers. Low variation in the NIs diameter and height has been found to lead to the uniform coalescence and it has been found very essential to achieve the device quality AlN layers.

AlGaN/GaN heterostructures (HS) were grown by MOCVD technique. AlGaN/GaN-HS growth was initiated with low temperature GaN nucleation layer on the sapphire substrate, followed by GaN buffer layer with the thickness of 1.5 µm and 65 nm thick AlGaN layer. The grown sample has
been cut into pieces of dimensions about 10 mm × 10 mm to perform irradiation. Irradiation effects on AlGaN/GaN-HS by 120 MeV silicon ion with the fluence of 5×10^{12} ions/cm^2 at room temperature and low temperature (liquid nitrogen temperature 77 K) were studied. The pristine and irradiated samples were characterized by HRXRD, AFM, Raman and photoluminescence spectroscopies. HRXRD studies have found to reveal there are no additional phase formations in the AlGaN/GaN-HS and no compositional changes in the Al_{0.2}Ga_{0.8}N layers after irradiation. FWHM of (102) reflection has been found to broaden for irradiated AlGaN/GaN-HS sample whereas the (002) FWHM values show only slight increase. The edge dislocations of AlGaN and GaN layers have been found to increase in AlGaN/GaN-HS samples upon irradiation. AFM images have been observed to show the formation of nano clusters after irradiation. The size of the clusters has been noted to increase in the low temperature irradiation and in turn rms surface roughness values were also found to increase. Predominant defect band emission has been found to show that the optical quality is highly deteriorated for the irradiated AlGaN/GaN-HS samples. Raman spectroscopy depicts only the allowed phonon modes for the backscattering geometry. That indicates the clusters formed on the surface of irradiated AlGaN/GaN-HS samples have only been in (002) orientation. The low temperature irradiation increases the surface roughness and defects of the AlGaN/GaN-HS, by the formation of big clusters and rapid quenching than the room temperature irradiation.

5.2 SUGGESTIONS FOR FUTURE WORK

To date, the external quantum efficiency of DUV-LEDs is less than 10%. The key part to improve the external quantum efficiency (EQE) lies in enhancing the quality of the AlGaN and AlN materials. The non-availability of III-nitrides substrate is the main reason for the higher dislocations density
in the order of $10^7 - 10^{11}$ cm$^{-2}$ and in turn the low quality of material. In this situation, the growth of high quality AlN layers certainly helps to increase the AlGaN based DUV-LEDs efficiency.

The utilization of the patterned sapphire substrates instead of normal substrate shall assist to improve the quality of AlN layers by suppressing the intrusion of threading dislocations into the active region. In addition to that, the extraction of light from the ‘c’ orientation has been found to be very difficult due to the anisotropic behavior of AlN. Hence, it is proposed to grow AlN on ‘a’ plane and ‘m’ plane substrates to improve the light extraction. Figure 5.1 shows the emission pattern of AlN.

**Figure 5.1 Emission pattern of AlN** [courtesy: Taniyasu et al 2010a]

Growth of p-type and n-type AlN and AlGaN layers needs to be optimized for better injection of carriers in the active region. Electrical characterization of AlN and high aluminum content AlGaN layer has to be
performed to understand the complete device characteristics. In addition to this, the growth of AlN and AlGaN layers at very high temperatures with respect to the carrier gas (H$_2$) flows need to be investigated for finding its effects on the crystalline quality.

Growth and fabrication of AlGaN/GaN based high electron mobility transistor (HEMT) with variation in the thickness of AlN interlayer has to be carried out. Implantation of silicon and magnesium ions has to be performed in the fabricated HEMT device to study the behavior of two dimensional electron gas (2DEG) and device characteristics with respect to ions, ion fluences and energy.