

The Influence of Nitrogen Ion Energy on the Quality of GaN Films Grown with Molecular Beam Epitaxy

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Since the growth of GaN using molecular beam epitaxy (MBE) occurs under metastable growth conditions, activated nitrogen is required to drive the forward synthesis reaction. In the process of exciting the nitrogen using a plasma or ion-beam source, species with large kinetic energies are generated. Impingement on the growth surface by these species can result in subsurface damage to the growing film, as well as an enhancement of the reverse decomposition reaction rate. In this study, we investigate the effect of the kinetic energy of the impinging nitrogen ions during growth on the resulting optical and structural properties of GaN films. Strong band-edge photoluminescence and cathodoluminescence are found when a kinetic energy of ~ 10 eV are used, while luminescence is not detectable when the kinetic energies exceeds 18 eV. Also, we find that the use of conductive SiC substrates results in more homogeneous luminescence than the use of insulating sapphire substrates. This is attributed to sample surface charging in the case of sapphire substrates and subsequent variation in the incident ion flux and kinetic energy across the growth surface. This study clearly shows that the quality of GaN films grown by MBE are presently limited by damage from the impingement of high energy species on the growth surface.

Key words: Activated nitrogen, GaN, molecular beam epitaxy (MBE), nitrogen ion energy

INTRODUCTION

Breakthroughs in the fabrication of GaN-based epitaxial device structures have led to the recent commercialization of high-efficiency blue light-emitting diodes. The critical step to this development came from Amano et al. who fabricated the first high-quality p-type epitaxial GaN films using Mg-doping.¹ In the same paper, the first GaN pn junction LED was also reported. The development of other blue LEDs quickly followed, and quantum efficiencies of the devices quickly surpassed those of their competitors in SiC and ZnSe.² In April 1994, Nichia Chemical announced the commercial development of a blue LED that boasted an external quantum efficiency of 2.7%.³

All of the described successes were developed with the same growth method, metalorganic chemical vapor deposition (MOCVD). In contrast, the fabrication of GaN device structures grown using molecular beam epitaxy (MBE) have not reached this high level of performance. The reason for this may come from the fact that GaN growth by MBE occurs under metastable growth conditions, while MOCVD growth of GaN occurs under thermodynamically stable conditions.⁴ Growth in which the compound is in thermodynamic equilibrium is also used for all conventional methods of epitaxial growth of group II-VI and III-V semiconductors.⁵

Despite operating under metastable conditions, MBE growth of GaN has had some success in growing high quality films. For example, GaN films have been made with mobilities which are higher than reported for MOCVD grown films.⁶ Also, p-type films have been

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recently produced by several different groups.^{6,7} There are also several important advantages of the MBE method over MOCVD. For example, films grown by MBE need no post-growth process such as low energy electron beam irradiation (LEEBI) or annealing for acceptor activation.^{6,7}

Metalorganic chemical vapor deposition-grown GaN films are typically grown in a hydrogen-containing environment and a post-growth process is necessary to remove the hydrogen passivation of acceptors.⁸ Since MBE growth is typically carried out without the presence of hydrogen-bearing species, MBE films do not require this additional processing step. Another advantage is that MBE growth allows much better control of layer thickness, and should make it the superior method to fabricate superlattices and multi-quantum well structures.

The motivation of this work is to understand and determine the conditions for optimum film growth in this new regime of metastable growth. In a recent paper,⁴ we outlined the dominant reaction kinetics and described the conditions under which GaN can be grown in this regime. This analysis can be used to understand how the growth parameters (such as the energy of the impinging species, the substrate temperature, and the ratio of Ga to N flux) affect the reaction. We briefly review the important points below.

Since the low background pressures used in conventional molecular beam epitaxy growth result in conditions in which Ga (l) and N₂ (g), or NH₃ (g) are the stable phase, additional energy must be supplied to the reaction to form GaN (s). In order to overcome this energetic barrier, a highly energetic form of nitrogen, such as N₂⁺ or N₂^{*} can be used to form GaN. The forward synthesis reaction of GaN is rate limited by the flux of impinging activated nitrogen species.

Since the molecular beam epitaxy process uses conditions in which GaN is not the stable phase, decomposition will also occur during the growth process. Because of the unusually large kinetic barrier of decomposition for GaN,⁹ this reaction can be kept to a rate significantly slower than the forward reaction rate; and growth in the metastable regime is, therefore, possible. Our analysis⁴ emphasized the importance of operating under conditions which minimize the reverse decomposition reaction in order to obtain high quality films.

Unfortunately, sources which produce activated nitrogen species such as electron cyclotron resonance (ECR) plasma sources, Kauffman ion sources, and microwave discharge sources also create significant concentrations of energetic ions, electrons, atoms, and/or molecules. Earlier work in the field has shown that the impinging flux of species with large kinetic energies can increase the rate of decomposition and create subsurface damage to the growing film.¹⁰ Brice et al.¹¹ have shown that subsurface damage results at ~3 times the semiconductor's cohesive energy, which for GaN is ~24 eV.

We chose a Kauffman ion source¹² for this study

because it provides a higher flux of near-monoenergetic nitrogen species at low kinetic energies than other commonly used sources such as the ECR source.¹³ For the Kauffman source, the minimum anode to cathode voltage required to produce a nitrogen plasma is 30 eV, resulting in a plasma potential of ~28 eV. Ion extraction to ground potential results in kinetic energies of ~+28V. Ions possessing this large a kinetic energy, clearly can cause enhanced decomposition, as well as damage to the lattice. In our experiments, we apply a DC bias to the substrate, thereby producing a retarding field which facilitates direct control of the incident ion kinetic energy.

Another variable which affects the reverse decomposition is the stoichiometry of growth. Schoonmaker et al.¹⁴ reported that the GaN decomposition reaction can be significantly enhanced from Ga-rich surfaces due to catalysis by metallic Ga. Since a number of investigators have studied the effect of this parameter on film growth,^{4,6,7,15} we will not include this in our report. For this experiment, an impingement ratio of 10:1 for N:Ga was chosen.

The focus of this study is on the effects of substrate bias and temperature on GaN film quality. Experiments were conducted on two different substrates, (0001) Al₂O₃ and (0001) n⁺ SiC. The motivation for the use of the two different substrates is that Al₂O₃ is insulating while SiC is conductive. We expect the bias to be applied uniformly over the SiC, and only locally near the metallic contacting clips for the Al₂O₃ due to sample charging of the insulating substrate.

EXPERIMENT

Ion-Beam MBE Chamber

The GaN films in this experiment were grown in a custom-built ion-beam MBE chamber. The chamber is equipped with three Knudsen cells to evaporate Ga, Al, and Mg. A quartz crystal rate monitor is used to measure the gallium evaporation rates, and a Faraday cup is used to measure the ion current. The substrate holder consists of a resistive element brazed in a molybdenum block. The chamber has load locking capabilities. Liquid-nitrogen cryopanel are used during growth. The main chamber is turbo-pumped and is fully bakeable. Ultimate base pressure is ~6 × 10⁻¹⁰ Torr.

The nitrogen source is a 3 cm Commonwealth Scientific Kauffman type ion gun. The discharge of the ion gun has been characterized with optical emission spectroscopy, and the major lines in the spectrum is a series of lines near 390 nm, which indicates the presence of N₂⁺. The nitrogen atomic lines at 745, 821, and 869 nm¹⁶ were not present. The Ga evaporation rate was typically set at 2000 Å/h.

1 cm x 1 cm (0001) SiC and (0001) Al₂O₃ were used. SiC substrates were degreased in boiling acetone followed by a rinse with ethyl alcohol and deionized water, and then dipped in HF. Sapphire substrates were cleaned with the same procedure without the HF dip. Indium was evaporated on the back of the

substrates and the substrates were then clamped to the heater block to ensure good thermal contact. For our experiments, a SiC and a Al₂O₃ substrate were used in each run.

Outgassing of the sample and substrates was performed at 400°C overnight, followed by 750°C for 30 min. A few monolayers of Ga were deposited in vacuum to initially cover the substrate surface in order to suppress the growth of Si₃N₄ on SiC. The nitrogen gas was then allowed to flow and the plasma struck. The filament of the ion source was degassed for 10 min before proceeding with growth. Growth was initiated with a 100Å buffer layer deposited at 500°C. The temperature was then ramped up to the growth temperature. After growth, samples were cooled in a nitrogen ambient of 10⁻³ Torr.

Characterization Tools

Photoluminescence (PL) was performed at 4K using an Omnichrome 30 mW HeCd laser ($\lambda = 325$ nm) as the excitation source. The emission from the sample is dispersed by a SPEX 1404 0.85 m double spectroctometer and detected by a Hamamatsu photomultiplier tube. A Stanford Research System SR540 chopper and SR530 lock-in amplifier were used to improve the signal-to-noise ratio. To measure the spatial homogeneity of the films, cathodoluminescence (CL) was used. The CL imaging system is based on a JEOL 35 CF electron microscope and Oxford liquid nitrogen cold stage. The emission from the sample is imaged using a parabolic mirror and is detected using a SPEX 1680B 0.22 m monochromator and a Hamamatsu photomultiplier tube. Structural information was obtained with a Siemens D5000 x-ray diffractometer with Cu-K α 1 tube, four-circle translational capability and four-bounce Ge monochromator. The thickness of each film was determined using a Dektak profilometer.

RESULTS AND DISCUSSION

Biasing Effects

To isolate the effect of the kinetic energy of the incident ions on film quality, films were grown with varying substrate biases while keeping all of the other growth parameters the same. A growth temperature of 675°C was used. The anode potential of the ion gun was set to 30V, and the potential difference between the cathode and the anode was set to 25V. The ion flux was maintained at 10¹⁴ cm⁻² s⁻¹.

Bias is applied directly to the molybdenum heater block which is in electrical contact with the molybdenum clamps. For the conductive SiC substrate, a uniform potential equal to the applied bias results across the substrate surface. The potential of the sapphire surface, on the other hand, will not be expected to be homogeneous across the sample and in the center of the sample can charge up to the floating potential at which there is no current flow. It will be near the applied bias only in the vicinity of the molybdenum clips.

Silicon Carbide Substrates

In Fig. 1, we compare the PL spectra taken from three different samples. Each is grown at a different bias: +0V, +9V, and +18V. A sample was also grown at +25V on a sapphire substrate, and no luminescence was detected. The luminescence near the band edge at 360 nm is attributed to bound exciton emission.¹⁷ This luminescence grows stronger with increasing substrate bias, which corresponds to decreasing incident ion energy. As seen in the figure, the film grown at +18V has strong luminescence near the band edge, while the other two films show luminescence 500 times less than the film grown at +18V. This is a clear indication that use of a substrate bias is improving the optical quality of the film. Therefore, we conclude that the high energy ions which bombard the surface of the unbiased films are resulting in damage which is detrimental to the optical properties of the film. The application of substrate bias to reduce the incident ion kinetic energy suppresses this effect. Also note that the midgap luminescence in all samples is very weak. In the sample grown with +18V substrate bias, the midgap luminescence is two orders of magnitude smaller than that the luminescence near the band edge.

From CL measurements, we monitor the spatial homogeneity of the films. All three films are reasonably homogeneous. Near the edges where the sample is clipped on the substrate, the luminescence on the sample biased at +18V is stronger by a factor of two. The luminescence of the films grown with +0, and +9 V, showed no significant variation across the surface.

Rocking curve and ϕ -scan x-ray measurements were also performed on these three films. The rocking curve was taken about the (0002) diffraction peak of

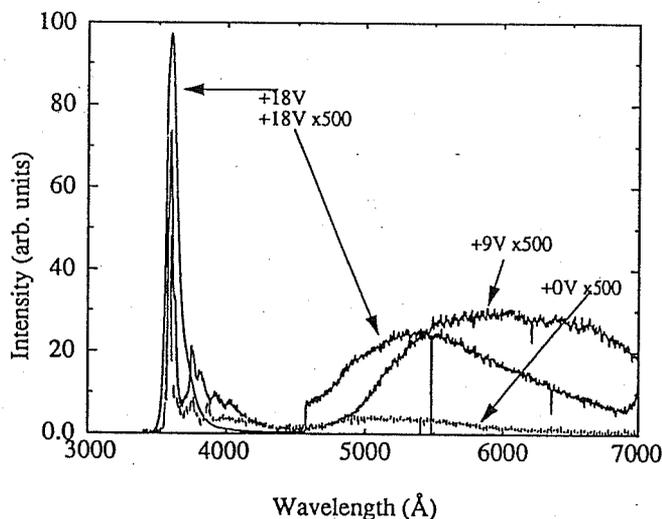


Fig. 1. Photoluminescence spectra of films grown on SiC with a substrate bias at 0V, +9V, and +18V. The measurement temperature is at 4K. Luminescence from the bound exciton grows stronger as the bias is increased and is dramatically larger for the +18V film. The spectra of the +0V and +9V films and the midgap emission from the +18V film are multiplied by a factor of 500.

Table I. Growth Rates of GaN Films Grown on SiC Substrates

Substrate Bias	Growth Rates ($\text{\AA}/\text{h}$)
0V	2000
+9V	1860
+18V	1710

Table II. Summary of X-ray Measurements on GaN Films Grown on SiC with a Substrate Growth Temperature of 675°C

Substrate Bias	FWHM	θ -Scan FWHM
0V	20 min	32 min
+9 V	24 min	42 min
+18 V	30 min	72 min
+18V (thick)	21 min	35 min

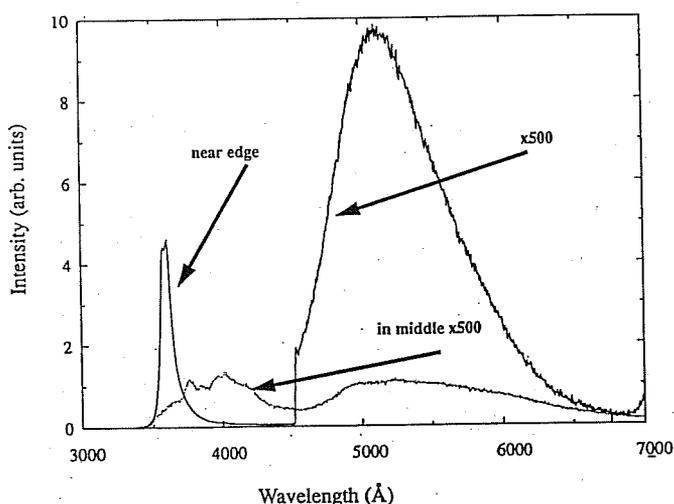


Fig. 2. Comparison of the PL spectra from two different regions of a GaN film grown on (0001) sapphire. Growth temperature of the film is 675°C , and the substrate is biased at +18V. Strong luminescence can be seen in the spectra taken near the edge of the film. The luminescence in the middle of the biased film is very weak and is similar to what is seen on an unbiased film. Data are taken at 4K.

GaN, and the ϕ -scans were made about the $(1\bar{1}02)$ off-axis peak. All three films showed sixfold symmetry in the ϕ -scan which indicates the films are single crystalline with a c-axis orientation. The rocking curve and ϕ -scan full width at half maximums (FWHMs) increased with substrate bias. The difference, however, can be attributed to the difference in thickness between the films. It is found that the growth rate is slower with increased substrate bias even though the Ga evaporation rate, and N_2^+ ion current were held constant. On SiC, the film grown with +18V substrate bias grew at a 15% slower rate than the film grown at +0V. The results are summarized in Table I.

To investigate the effect of thickness on the x-ray measurements, an additional film was grown with +18V substrate bias and was made the same thickness as the previously grown +0V film. The FWHMs

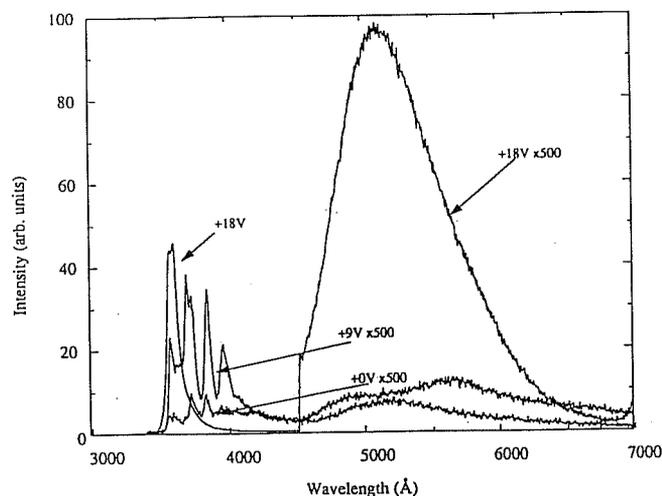


Fig. 3. Comparison of luminescence of three films grown on sapphire with substrate bias of 0V, +9V, and +18V.

of this film were comparable to the film grown at +0V. The data is summarized in Table II.

Sapphire Substrates

In contrast to the luminescence from the SiC samples, the luminescence from films grown on sapphire was not laterally uniform. Near the edges, where the clips touch the sample, strong luminescence was found. Away from the edges, there is no detectable luminescence. This result is not surprising because the potential on the sapphire is nonuniform due to surface charging effects. The inhomogeneity of the luminescence indicates that the bias is strongly affecting the optical properties of the GaN films. Figure 2 contrasts the PL spectra obtained from two regions in the film. The strong luminescence is observed for regions near the edge, and the weak luminescence is observed for regions in the middle of that same film.

The photoluminescence near the edge of the films grown on sapphire, as illustrated in Fig. 3, shows similar results to those seen on SiC. Increasing luminescence is found for bound exciton emission with increasing bias. And again, the film biased at +18V shows a dramatic improvement over those biased at +9V or +0V. Note the similarities between the spectra taken at +0 V in Fig. 3 and the spectra taken in the middle of the sample in Fig. 2. The similarity suggests that the potential of the surface in the middle of the films is not at the applied bias.

An important point to note is that the ratio between midgap and bound exciton emission is larger for the film grown on sapphire than the film on SiC. Comparing the two films grown at +18V, the film grown on SiC has a exciton/midgap ratio of 2500:1 while the film grown on sapphire has only a 250:1 ratio. The power of the laser was kept constant for these experiments so the ratios are directly comparable. It is important to keep the laser power constant because, as Moustakas has noted,¹⁸ the ratio of midgap/exciton peaks is a function of the pumping laser power due to saturation of the midgap emission.

To further illustrate the difference in lateral homogeneity between films grown on SiC and sapphire, cathodoluminescence was performed. The CL spectra from films on sapphire are found to be much less uniform than that on SiC. For the films grown on sapphire, the luminescence from the middle of the samples are three orders of magnitude less than that on the edge where the films were clipped. The width of these high-luminescence regions is on the order of 100 μm . On the other hand, the luminescence on the edge of the SiC films is found to be at most a factor of two larger than that in the middle.

In order to further test our conclusions, we seek another method to reduce the energy of impinging ions at the growth surface. By increasing our gas pressure to 15 mT in combination with substrate bias, the kinetic energy of the impinging species is expected to be significantly reduced by scattering (the nitrogen mean-free-path is ~ 1 mm at this pressure). The resulting film on sapphire was found to be homogenous in luminescence across the sample. The structural properties also showed that high quality films were made with x-ray rocking curve FWHM of 11 min for the GaN (0002) peak.

Rocking curve and ϕ -scan x-ray measurements were also made on the films grown on sapphire substrates. The measurements were taken in the middle of the sample. The rocking curve was taken about the (0002) GaN peak, and the ϕ -scan was taken about the (1 $\bar{1}$ 02) off-axis peak. The films on sapphire showed sixfold symmetry in the ϕ -scan indicating a single crystal oriented with the c-axis perpendicular to the substrate. Just as in the SiC case, we find the rocking curve and ϕ -scan FWHMs increase with applied bias. Again, this result can be explained by the difference in thickness of the biased and unbiased films. Similar to the discussed results on SiC, biased films on sapphire have a slower growth rate than the unbiased films. To compensate for this difference a film was grown with +18V substrate bias with the same thickness as the +0V film, and the x-ray FWHM of the scans were found to be similar. The results are summarized in Table III.

Interestingly enough, the films grown on sapphire have comparable or better crystallinity than the films on SiC. In fact, comparing the two films grown with +18V substrate bias, the film on sapphire has half the width of the film grown on SiC. This result is surprising because the lattice mismatch for GaN on SiC is only 2%, while the mismatch between GaN and sapphire is 14%. One would speculate that the SiC would be a much better substrate to grow GaN films on because of the more favorable lattice matching. A possible explanation for why the films on SiC are of poorer quality than those on sapphire is that nonepitaxial Si_3N_4 film is formed at the film-substrate interface. This indicates our procedure of evaporating the Ga metal with the nitrogen gas off is not completely successful at suppressing the growth of nonepitaxial Si_3N_4 , presumably due to incomplete coverage of the surface due to clustering of metallic Ga.

Discussion of Biasing Results

In the following section, we discuss the influence of the deposited film on the homogeneity of the potential at the growth surface when biasing is applied. The resistivity of an undoped film on sapphire was measured to be 1000 $\Omega\text{-cm}$ at 700°C. Using this as an estimate of the resistance, we can calculate the voltage drop that we would see across the film on an insulating and a conducting substrate. For a 1000Å film, for example, the resistance between the middle of the film and the clip is approximately 25 M Ω .

In the case of a conducting substrate, the current passes only through the thickness of the GaN film. Again, taking the thickness to be 1000Å, and assuming we have an ohmic contact between the SiC and the GaN film, the resistance between the surface of the film and film-substrate interface is negligible, 0.01 Ω .

The current from the ion flux over the area of the substrate is approximately 100 μA . For this current, we get a negligible voltage drop (~ 1 μV) across the thickness of the film. Laterally, however, the film can support the entire voltage drop of the substrate bias. Therefore, in the SiC case a homogeneous potential results across the sample, but in the sapphire case there is a large potential drop between the middle and the edge of the surface.

Growth Temperature Effects

To investigate the effects of growth temperature on GaN quality, films were grown at 600, 675, and 750°C on a SiC substrate. Previous experiments have found

Table III. Summary of X-ray Measurements of GaN Films Grown on Sapphire with a Substrate Growth Temperature of 675°C

Substrate Bias	FWHM	θ -Scan FWHM
0V	11 min	26 min
+9V	24 min	42 min
+18 V	34 min	72 min
+18V (thick)	12 min	26 min

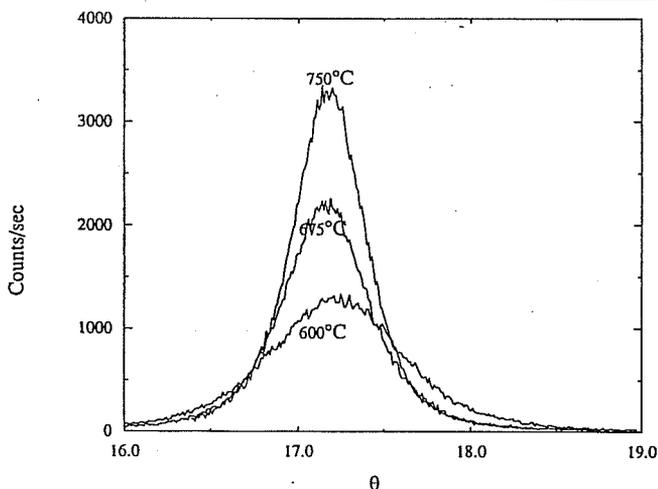


Fig. 4. Rocking curves of (0002) GaN peak for films grown on SiC (0001). θ is calibrated by the (0006) SiC peak.

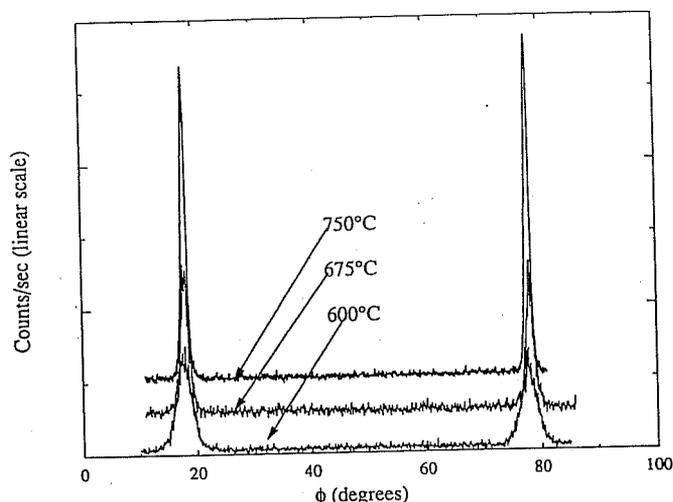


Fig. 5. ϕ -scans of $(1\bar{1}02)$ peak of GaN films grown on SiC. As the growth temperature increases from 600 to 750°C, steady improvement in the crystalline quality is evident.

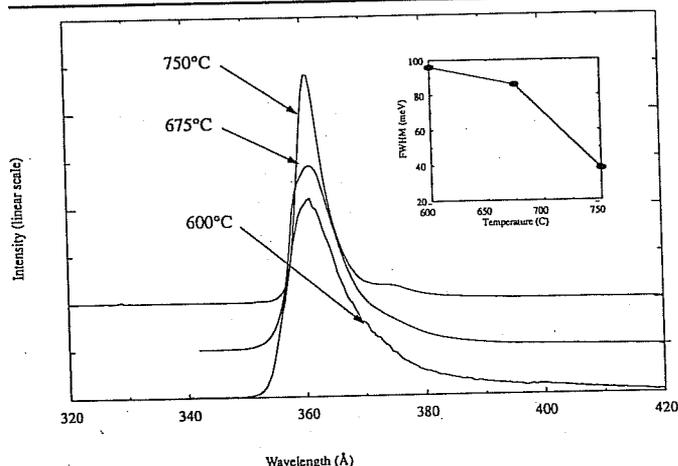


Fig. 6. Photoluminescence spectra of GaN films grown at 600, 675, and 750°C. The bound exciton peaks of the three films are all centered at 3600Å. The three spectra are vertically offset from each other. In the inset, the FWHMs of the exciton emission peaks are plotted as a function of temperature.

that films grown with a substrate temperature below 600°C have (0002) rocking curve widths greater than 50 min and no detectable peaks are found in the ϕ -scan; indicating the material is not single crystalline.

The x-ray rocking curves of the (0002) GaN peak for films grown at 600, 675, and 750°C on SiC are compared in Fig. 4. The three films are all grown with a substrate bias of +18V and the thicknesses of the films are all within 5% of 6000Å. The rocking curves were calibrated to the (0006) peak of the SiC substrate, which is set to $\theta = 17.79$ degrees, corresponding to a c-axis lattice constant of 15.12Å. As the temperature increases from 600 to 750°C, the FWHM of the GaN (0002) peak decreases from 52 to 28 min and the intensity increases by a factor of three.

Similarly, the widths of the peaks in the ϕ -scan also improve with increasing temperature. Figure 5 shows the ϕ -scans of the $(1\bar{1}02)$ GaN peak of the samples described in Fig. 4. All three films show peaks every 60 degrees, indicating that all three films are single

crystalline. The FWHM improves significantly as the temperature is increased from 600 to 750 C. From the x-ray data, it is clear that the epitaxy is still improving with increasing temperature. From this data, we conclude the point of significant GaN thermal decomposition has not yet been reached.

Temperature also has a small, but measurable effect, on the photoluminescence of the films. The PL spectra of the above described films were taken at 4K, and the plots appear in Fig. 6. The three spectra in this figure have been vertically offset from each other for clarity and the exciton peaks are all centered at 3600Å. Each of these films show midgap luminescence three orders of magnitude weaker than the exciton emission. The luminescence narrows as the growth temperature increases. The FWHMs of the exciton peak are plotted in the inset of Fig. 6. From Fig. 6, it is clear that higher temperature improves the optical quality of the film.

SUMMARY

We conclude that by applying a bias to the substrate during film growth, we can lower the incident ion kinetic energy which leads to significantly improved optical properties of the grown epitaxial GaN films. Using the Kauffman source, it is necessary to use a conductive substrate to obtain uniform quality across the film. The crystallinity, as determined by x-ray diffraction, however, is not significantly improved when substrate bias is applied. This suggests that the damage caused by high energy ions is of a point defect nature because such a defect would have a smaller effect on the widths of the x-ray diffraction peaks. Point defects, however, can be very harmful to the optical properties, as evidenced by the data in section on biasing effects. From the data taken in the 600–750°C range, it is clear that growth at 750°C provides superior material than the other temperatures. Both the structural properties, and the optical properties are improved in the high temperature films.

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