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XRD and photoluminescence measurements of GaN grown on dome shaped patterned sapphire with different NH_3 flow rates

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Abstract

The aim of the study is to understand the effects of NH_3 flow rate in the initial part of high temperature (HT) GaN growth on structural and optical characteristics of the HT-GaN layer grown on dome shaped sapphire susbtrate by Metal Organic Chemical Vapor Deposition (MOCVD) system. High resolution x-ray diffraction (HRXRD) and photoluminescence (PL) measurements were performed to characterization the growing GaN epilayer. It is observed that the using of different NH_3 flow rate in the initial part of HT-GaN growth has an effect on both full-width at half-maximum (FWHM) values obtained from HRXRD results and intensities of yellow luminescence peaks. It is seen that the FWHM values obtained from the symmetric (00.2) omega scan increased as the NH_3 flow rates in the initial part of HT-GaN growth increased. It is demonstrated that the intensities of yellow luminescence peaks are very sensitive to NH_3 flow rates in the initial part of HT-GaN growth.

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1. Introduction

III-Nitride compound semiconductors (GaN, InN, AlN, and their alloys) have attracted great attention and achieved incredible progress in recent years. The main underlying reason is that GaN and its alloys with AlN and InN have huge application area for optoelectronic device applications (light emitting diode (LEDs), laser diodes and hiph power and high temperature electronics) [1-6]. MOCVD system is currently the most widely used technology to produce GaN based LEDs. Since the first high-brightness GaN-based blue LED was achieved on the sapphire substrate, researches to obtain LED with high power and high efficiency are still a hot topic area. However, further development in LED is hampered due to two main hindrances. First, GaN has a high crystal defect density because of the large lattice mismatch between GaN and sapphire resulting in impacts the internal quantum efficiency (IQE) of LEDs [7-9]. Second, the light extraction efficiency (LEE) is low due to GaN/sapphire interface [10, 11]. There are plenty of MOCVD growth studies in the literature to investigate the effect of growth parameters on defect density in GaN on flat sapphire substrate. In these studies, the effect of thickness [12], growth temperature [13], reactor pressure [14], annealing [15], total flow [16], V/III ratio [17], and many other growth parameters of LT (low temperature) GaN nucleation layer and HT (high temperature) GaN layer were deeply investigated. While patterned sapphire substrate (PSS) has proven major advantageous for improving the efficiency of LED, PSS also contributes one step epitaxial growth tecnologhy to obtain the lateral growth of GaN which is important to overcome the disadvanteges of the two step epitaxial growth [18]. In this regard, using PSS instead of the flat sapphire substrate can improve the epitaxial lateral overgrowth (ELOG) mode and consequently provide to obtain high quality GaN. The crystal defects in the GaN layer can be find as non-radiative recombination centers and therefore decrease some properties such as carrier lifetime and radiative recombination efficiency which are important as regards the efficiency of LEDs [19, 20]. NH_3 flow rate which defines the V/III ratio is also one of the important growth parameters and affects the quality of GaN epilayer. NH_3 flow rate or V/III ratio has important role during transition from 2D to 3D growth. In literature, the effect of V/III ratio or NH_3 flow rate is done for during low temperature GaN (LT-GaN) growth or high temperature GaN (HT-GaN) by using flat sapphire substrate.

In the current study, NH_3 flow rates in the initial part of the HT GaN growth where the transition from 2D to 3D growth mode take place have been changed to improve the quality of the GaN epilayer grown on the dome shaped sapphire substrate. The impact of

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*Corresponding author. *email address: ialtuntas@cumhuriyet.edu.tr* different NH_3 flow rates in the initial stage of the HT-GaN layer growth on structural and optical properties are studied using HRXRD system, and PL spectroscopy, respectively.

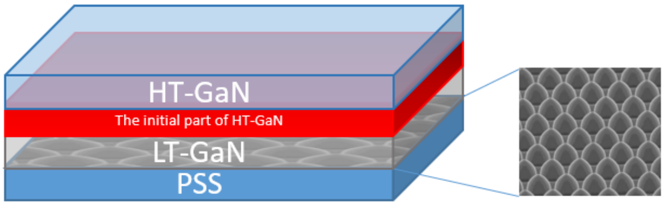
2. Experimental

A series GaN epitaxial layers were grown on dome shaped patterned sapphire substrates by AIXTRON 200/4 RF-S MOCVD system with the horizontal flow. The high purity ammonia (NH_3) and trimethylgallium (TMGa) were used for N and Ga sources, respectively, to grow epitaxial GaN layer. High purity H_2 gas was used as the carrier. All samples were deposited with the same growth conditions except for NH_3 flow rates in the initial stage of HT-GaN growth where the 3D to 2D growth transition take place. Before the growth, the dome shaped patterned sapphire substrates were applied the high temperature desorption (1080 °C) for 10 mins at 100 mbar reactor pressure under H_2 ambient to remove any remaining contaminants from the surface of sapphire substrates. GaN grown at low temperature which is called the nucleation layer was grown at 200 mbar and 480 °C. After nucleation layer growth, the growth temperature was incrased to 1030 °C and reactor pressure was fixed to 200 mbar for HT-GaN growth. The GaN layer grown at higher temperature was included 2 parts with different NH_3 flow rates. While the initial part of HT-GaN was grown for 60 mins with different NH_3 flow rates, remaining part of HT-GaN was grown for 40 mins. NH_3 flow rates in the initial part of HT-GaN were changed as 1250, 1000 and 750 sccm while NH_3 flow rate at second part of HT-GaN growth was kept constant at 1250 sccm. These samples were labeled as S1, S2, and S3. Table I summarizes the NH_3 flow rates in the initial part of HT-GaN growth.

Table 1. The NH_3 flow rates in the initial part of HT-GaN for S1, S2, and S3.

Sample Name	NH_3 flow rates in the initial part of HT-GaN (sccm)
S1	1250
S2	1000
S3	750

Figure 1 demostrates the schematic representation of growth structure and used dome shaped patterned substrate. NH_3 flow rates in the red layer (initial part of HT-GaN) were adjusted as 1250, 1000, and 750 sccm for S1, S2, and S3, respectively.



a) b)

Figure 1. a) the growth structure used to obtain high quality GaN epilayer and b) the dome shaped patterned substrate.

HRXRD and room temperature (RT) photoluminescence measurements were done for structural and optical chracterization of these samples, respectively. HRXRD measurements were used to obtain the full-width at half-maximum (FWHM) values to investigate the effect of NH_3 flow rates in the initial part of HT-GaN growth on the structural quality. RT-PL measurements were carried out by the 325 nm HeCd laser which focused on the 10x objective lens for different laser powers. The power of the laser with 100mW was adjusted as 0.1, 1, 5, and 10% by using ND filters. The signal during the photoluminescence measurement was detected by the CCD detector and used 300 gr/mm 500nm blaze grating.

3. Results and Discussion

HRXRD is a commonly utilized method to investigate the structural properties of epitaxial layers for optimization of growth process [21-23]. The rocking curve omega scans for (00.2) and (10.2) planes were done and their FWHM values were used to take information about crystal quality of layer. Figure 2 gives rocking curves omega scans for (00.2) and (10.2) planes and also shows the change of FWHM values obtained from symmetrical and asymmetrical scans with chancing NH_3 flow rates in the initial part of HT-GaN growth.

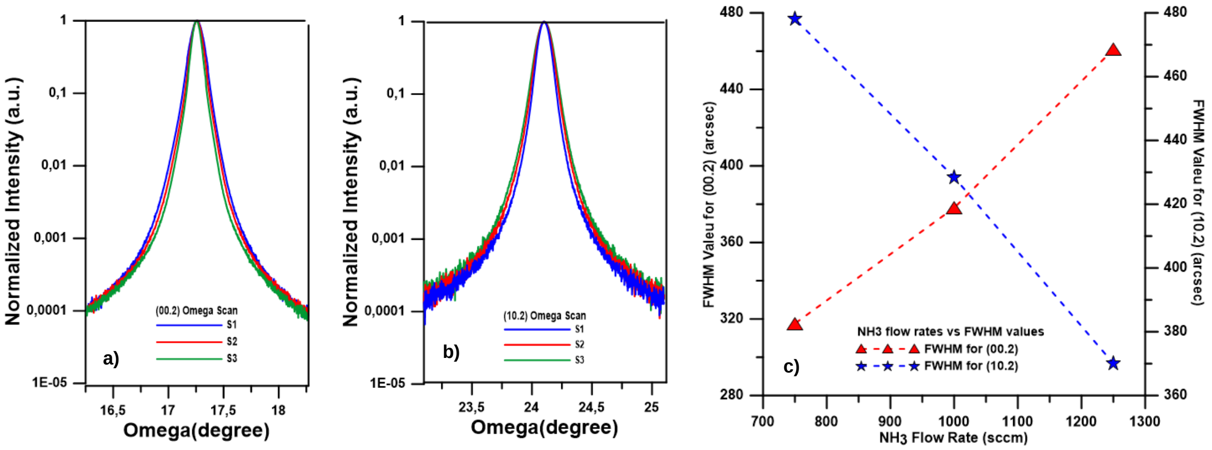


Figure 2. Omega scans for (a) (00.2) and (b) (10.2) planes and (c) the variation of FWHM values with the increase of NH_3 flow rates in the initial part of HT-GaN growth.

The basic aim of using different NH_3 flow rates in the initial part of HT-GaN growth is to optimize the NH_3 flow rates and to minimize the FWHM values obtained from symmetric and asymmetric rocking curve measurement for improving the crystalline quality. It is observed that although there is a linearly increasing dependency between the FWHM values obtained from (00.2) omega measurement and NH_3 flow rates, there is a linearly decreasing dependency between FWHM values obtained from (10.2) omega measurement and NH_3 flow rates. According to the results of HRXRD measurements, the desired growth condition can be determined in terms of the variation of FWHM values in both symmetric and asymmetric rocking curve measurements.

PL measurement is the most common, non-destructive, and fast optical characterization technique to understand the optical properties of structures [24].

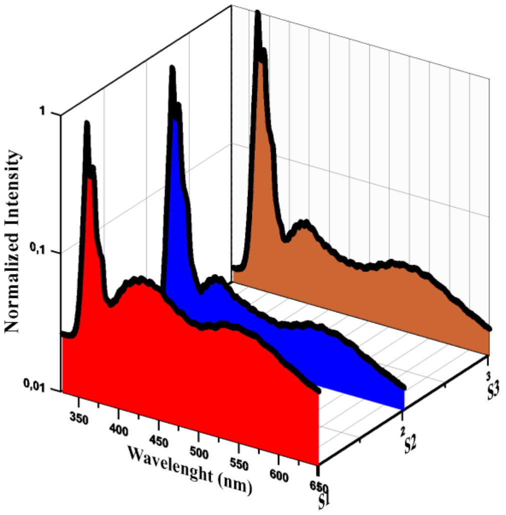
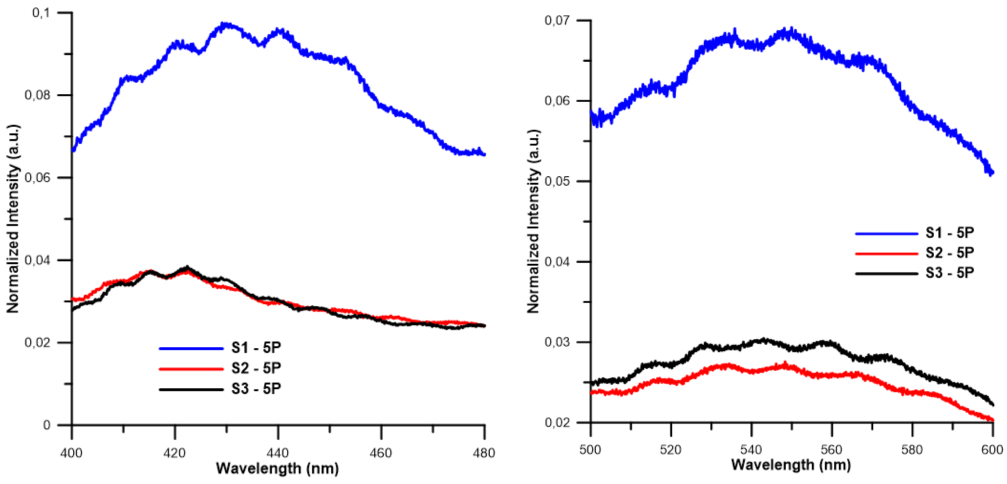


Figure 3. The normalized photoluminescence spectra for S1, S2, and S3 with different NH_3 flow rates in the initial part of HT-GaN growth.

For this reason, photololuminesce measurements were done to understand the effect of different NH_3 flow rates in the initial part of HT-GaN on the quality of the GaN epilayer. Figure 3 shows the normalized PL spectra with 330-650 nm range at RT for S1, S2, and S3 by laser power adjusted 5 %.

Each PL spectra was obtained from the center of the samples and adjusted laser power with 5% (5P). The PL spectra for S1, S2, and S3 have three main peaks; (1) high intensity and narrow PL band called near band edge luminescence at ~361 nm and (2) and (3) low intensity and broad PL band called yellow luminescences at ~ 430 and 550 nm. In GaN crystal, the band edge luminescence is caused by the transition from the conduction band to the valance band while yellow luminescences are caused by point defects which are sources of impurity and growth conditions [25]. Point defects have a very important role in the electrical and optical properties of GaN based materials/devices. However, the source of point defects in this semiconductor is still under investigation [25]. Figure 4 demostrates the comparison of PL peak emissions at ~ 430 and 550 nm for S1, S2, and S3. Fluctuations in the signal are caused by Fabry-Pérot oscillations (FPO) of the sapphire-GaN-air space [26].



a) b)

Figure 4. The comparison of PL spectra emissions at ~ 430 (a) and 550 nm (b) for S1, S2, and S3

It was observed that PL peak emission wavelength changed at around 430 nm while PL peak emission wavelength remained constant at around 550 nm for different NH_3 flow rates in the initial part of HT-GaN growth. The reason for this shift at around 430 nm is not completely clear but might be due to more yellow luminescence from deep centers depending on NH_3 flow rates. Generally, a common way to compare the quality of GaN epilayer is that the intensity ratio (I_{NBE}/I_{YL}) between near band edge peak and yellow luminescence peak is found [27]. For this purpose, the I_{NBE}/I_{YL} ratio was plotted against NH_3 flow rates used in the initial part of the HT-GaN growth (Figure 5).

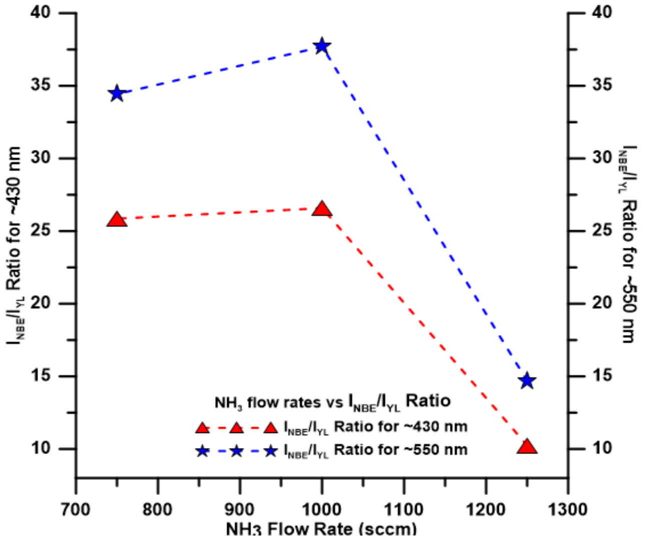


Figure 5. The variation of the I_{NBE}/I_{YL} ratios with NH_3 flow rates for 430 (red) and 550 (blue) nm.

PL measurement result and obtained the I_{NBE}/I_{YL} ratios indicate that the S2 can be chosen to optimize the growth condition. Also the effect of the using different laser powers on intensities of near band edge peak, and yellow luminescence peaks in the PL emission spectra of S2 was investigated. The PL emission spectra with 300-700 nm range were given for different laser powers in Figure 6. With increasing laser power, the NBE band intensity increased, what can be explained by reduction of the depletion region width [28]. It can be said that the intensities of yellow luminescence peaks become more pronounced with increasing laser power and also the intensity of the band-edge peak gradually increases.

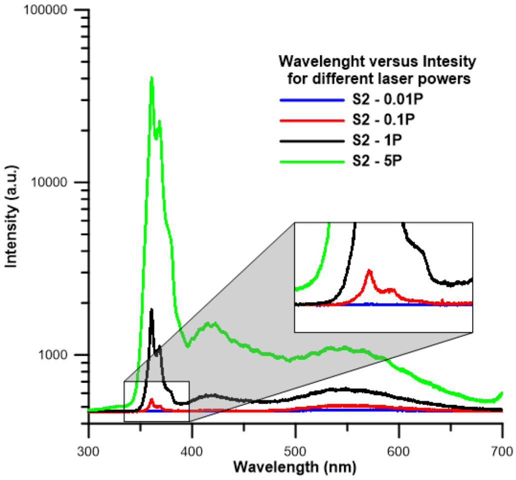


Figure 6. The PL emission spectra with 300-700 nm range obtained from excited by different laser powers for S2.

4. Conclusion

In summary, the impact of NH_3 flow rates in the initial part of HT-GaN growth is investigated on structural and optical properties of GaN epilayer grown on dome shaped sapphire substrate by using HRXRD and PL systems. The symmetric (00.2) and asymmetric (10.2) rocking curve measurements were done to obtain the FWHM values. The change of FWHM values obtained from both symmetric and asymmetric scans was plotted depending on the changing NH_3 flow rates. It is seen that there is a linear relation between the FWHM values obtained from (00.2) omega measurement and NH_3 flow rates in the initial part of HT-GaN growth. PL emission spectra with 300-700 nm range for S1, S2, and S3 were obtained to compare the effect of NH_3 flow rates. The variation of the intensity ratio (I_{NBE}/I_{YL}) with NH_3 flow rates was plotted to investigate the effect on the quality of the GaN epilayer. It is seen that the S2 has a higher I_{NBE}/I_{YL} ratio compared to the other samples.

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Conflicts of interest

The authors state that did not have conflict of interests.

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