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Optimization of the growth of GaN epitaxial layers in an indigenously developed MOVPE system

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Abstract. Growth of GaN epitaxial layers is optimized in an indigenously developed nitride metal organic vapour phase epitaxy (MOVPE) system. The motivation of present work is to develop a simple MOVPE reactor which is though affordable but can deliver GaN epilayers of desired quality. The design and fabrication methodology of nitride MOVPE system is briefly discussed. MOVPE growth of GaN epilayers is carried out through a two-step growth process where influence of the growth parameters of low temperature buffer layer on the crystalline properties of high temperature GaN epilayer is discussed in detail. Optimum values of the growth temperature, annealing duration and thickness of GaN buffer layer are achieved. High resolution X-ray diffraction measurements (HRXRD) confirmed that the crystalline quality of GaN epitaxial layers is reasonable. A good surface morphology of the optimized sample is strongly corroborated by the results obtained from photoluminescence (PL) and HRXRD techniques. Usefulness of a systematic Williamson-Hall analysis in the optimization of growth of GaN epitaxial layers is demonstrated where a high value of lateral coherence length of GaN buffer layer is found to be a key parameter. Such an optimization process leads to the good crystalline quality of GaN epitaxial layers with low dislocation density. It is found that the high temperature GaN epilayers grown on the optimized GaN buffer layer are compressively stressed which is also revealed by the PL measurements.

Keywords. MOVPE; GaN; buffer layer; HRXRD; PL.

1. Introduction

Considerable progress in the research and development of Gallium nitride (GaN) and its related ternary alloys involving Al and In is mainly driven by the improvement in the crystalline quality of nitride epitaxial layers. Nitride epitaxial layers are essential for the realization of high performance optoelectronic devices and are grown either by metalorganic vapour phase epitaxy (MOVPE) or molecular beam epitaxy (MBE) technique [1–5]. One of the major challenges in the growth of high quality GaN is related with the lack of suitable lattice matched substrate. In view of this, GaN epitaxial layers are usually grown on various foreign substrates like Si, SiC and Sapphire etc. [6–8]. Recently native GaN substrates are also being used by a few researchers for the growth of high crystalline quality nitride epilayers [1, 9, 10]. However, foreign substrates like Si, SiC and Sapphire still make the primary choice for the development of various nitride based semiconductor devices [1, 5, 11]. In particular, Sapphire has played a pivotal role in the development of high efficiency light emitters operating in the ultraviolet-visible range. Owing to a large lattice mismatch and high contrast in the values of thermal expansion coefficients of GaN and Sapphire, the growth of GaN epilayer on Sapphire usually takes place in two steps [12, 13]. In general, a thin GaN or AlN buffer layer is grown at low temperature before the high temperature growth of a thick GaN epitaxial layer [14, 15]. The buffer layer consists of small crystalline grains that act as nucleation centres for the growth of high temperature GaN epilayers. At high temperature, a large density of small nucleation centres is replaced by a small number of bigger islands which ultimately coalesce to form the required epilayer. In that sense, a low temperature (LT) grown buffer layer promotes the lateral growth of main epi-layer and also helps in lowering the dislocation density. Here, one of the

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most tedious issues is to find an optimum set of growth parameters for the LT buffer layer. Considerable progress has already been made where impact of a particular growth parameter on the layer quality is reported by different groups. From the early studies, it was found that growth of LT buffer makes the layer amorphous, which covers the entire substrate surface uniformly upon heating to the high temperature before the growth of thick GaN layer [13, 14]. The buffer layer actually crystallizes upon heating and provides excellent surface for further epitaxial growth. Hence, it is well known that the optimization of LT buffer and high temperature GaN layer is essential for achieving high quality GaN layer. Although a consensus about the optimize set of parameters seems to exist for the growth of GaN epilayer, still conflicting reports keep appearing in literature. For example, contrary to the common belief that the requirement of a LT buffer layer is mandatory, a few researchers tend to embark on the high temperature (HT) grown buffer layer [16]. At the same time, a wide range of growth parameters have been reported by several groups, for example nitridation [12, 13], V/III ratio, growth temperature [17, 18], reactor pressure [19, 20], growth rate [21] and the layer thickness [22, 23] parameters vary over a wide range. It seems that though many critical issues related to the MOVPE growth of GaN epitaxial layers appears to be well understood, in reality a lot more is yet to be learnt. In order to understand the role of LT-buffer layer, researchers have even tried to revisit their own concepts due to the availability of new in-situ probe in MOVPE growth [24]. Moreover, it seems that the optimization of growth recipe varies from one reactor to another and one has to find his own set of growth parameters to deliver the best quality of layers from a given MOVPE system.

Even for the state-of-the-art MOVPE reactors, most of the growth parameters are mandatory to be fine-tuned before the desired quality is achieved. Most often the set of growth parameters is specific to a particular reactor design/geometry and the growth recipes cannot be easily transferred from one reactor to another. Moreover, such MOVPE reactors are extremely expensive and are beyond the reach of common researchers. It therefore provides scope for the development of a simple MOVPE reactor which is affordable and can deliver the layers of desired quality. In this article, we report the indigenous development of a nitride MOVPE reactor, which is the main objective of present work. First, the layout of MOVPE system and the fabrication methodology is discussed in brief. Next, the growth of GaN epitaxial layers by indigenously developed nitride MOVPE system is discussed. In particular, the importance of three critical parameters i.e. the thickness of LT buffer, annealing time and temperature of LT buffer is studied for the optimization of growth of GaN epilayer at high temperature. Structural and optical properties of LT buffer as well as HT GaN layer are investigated using high resolution x-ray diffraction (HRXRD) and photoluminescence (PL) techniques. Surface morphology of MOVPE grown GaN epitaxial layers is studied by Field Emission Scanning Electron Microscopy (FESEM) and Atomic Force Microscopy (AFM) techniques.

2. Design of Nitride MOVPE system

The schematic diagram of indigenously developed nitride MOVPE system is shown in figure 1. Standard precursors like tri-methyl Gallium (TMGa), tri-methyl Aluminium (TMAl), tri-methyl Indium (TMIn), and Ammonia are used for the growth of GaN. For the doping purposes, diluted Silane (2% in Hydrogen) is also included. The design, fabrication, assembly and testing of nitride MOVPE system is discussed in this section.

As shown in figure 1, Nitride MOVPE system can be divided into the following sub-systems:

1. Gas supply and Mixing System (GMS) including the gas cabinets
2. Stainless steel reactor with a gas injection shower head and a high temperature ($\sim$1100 °C) heater
3. Vacuum system
4. Scrubber unit
5. Ventilation system
6. Control circuitry including computer control of the entire MOVPE system

Photographs of the developed nitride MOVPE system, GMS unit and the MOVPE reactor are shown in figure 2. Fabrication of the system was carried out under a clean tent of class 100 specifications in order to avoid any contamination during the fabrication. All the openable joints are made with the face sealed fitting whereas the permanent joints are made by using an orbital welding machine. In order to ensure the cleanliness of tube joints, high purity Argon gas is continuously flown during the welding process.

GMS of nitride MOVPE provides the various precursors in desired proportions and sequence to the MOVPE reactor depending on the requirement of a particular semiconductor material to be grown. It is an ultra-high purity (UHP) gas handling system where all the components are carefully selected to ensure their compatibility with UHP gases. No impurities shall be generated in these components. Further, a very high leak tightness is mandatory due to the corrosive and toxic nature of precursors. It is also required for minimizing the impurity incorporation in the grown layer. All the components of GMS like mass flow controllers, pressure controllers, valves, regulators, tees, elbows and bends are made of 316L stainless steel and are chosen to be of appropriate grade in order to meet the epilayer quality. An indigenously designed shower head of 2-inch diameter is used to feed the gaseous precursors to the nitride MOVPE reactor. A very important criteria of the gas injection is that there should be a minimal premixing between the group III

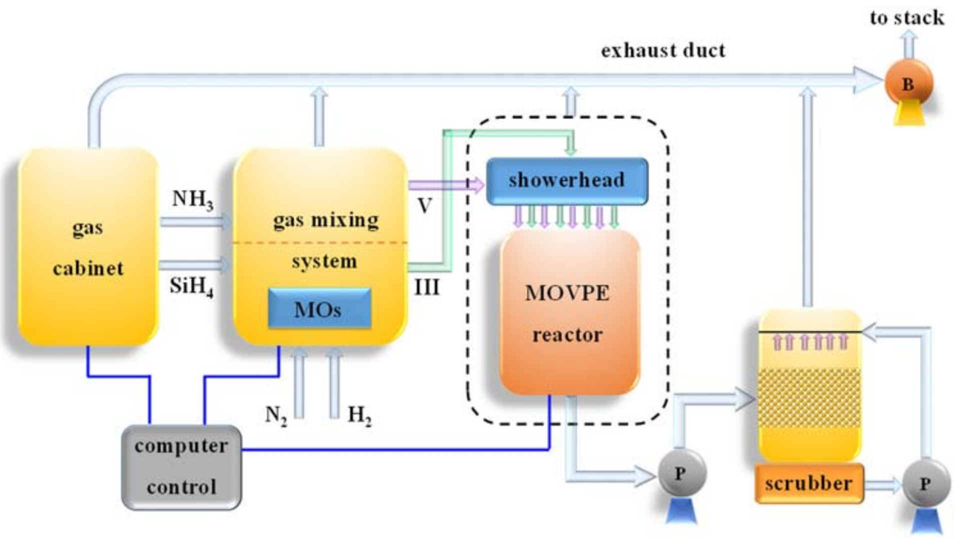

Figure 1. Schematic diagram of indigenously developed nitride MOVPE system.

Figure 2. Real photographs of (**a**) indigenously developed nitride MOVPE system, (**b**) gas mixing system, and (**c**) MOVPE reactor.

and group V precursors. In our design, MO sources enter the shower head from the top flange and are fed into the reactor through 25 holes of 1 mm dia. On the other hand, Ammonia gas enters the shower head from the side plenum and is fed into the reactor through 56 holes of 1 mm dia. In the shower head, these holes are arranged in such a manner that every MO hole is surrounded by four holes of Ammonia. It therefore supports a uniform gas mixing inside the MOVPE reactor. Premixing of the precursors is thus minimized by the separate injection of group III and group V precursors. The material of the shower head is stainless steel of SS316L grade. The most important aspect of the showerhead design is the fact that the complete assembly is fabricated from a single stainless steel block. Hence, no weld joints are exposed to the corrosive gases during the reactor operation. Another important concept in MOVPE reactor design is related to the uniform mixing and laminar flow of gases. Recirculation of gases inside the MOVPE reactor needs to be minimized in order to avoid memory effects and also to minimize the resident time of un-reacted precursors. This is particularly important when one aims to achieve sharp interfaces during the hetero epitaxial growth. Such issues are taken care of by choosing vertical reactor geometry along with the showerhead. The vertical reactor geometry with gas injection from the top and exhaust port at the bottom helps in maintaining a streamline flow of gaseous precursors. Since the showerhead is mounted on the top flange of the reactor, the buoyancy induced recirculation of gases can be minimized by varying the total gas flow in the reactor up to certain extent. In our design, Hydrogen and Nitrogen are used as the carrier and purge gases respectively. It is ensured that the purity of H_2 and N_2 gases is of the order of 99.999% (5N pure). However, in order to grow epilayer of excellent crystalline quality, much higher purity ($\geq$7N pure) is required. This is achieved by using getter based purifiers in the indigenously developed MOVPE system. Ultra-high purity H_2 gas is therefore used to transport the MO precursors into the reactor. For achieving high temperature required for the growth of GaN, an indigenously designed high temperature resistive heater capable of reaching 1100 °C is also included. The heater and substrate holder are made of high purity Molybdenum metal. Diameter of substrate holder and heater is kept 40 and 35 mm respectively. The substrate holder is kept 2 mm above the heating element. Growth temperature is measured by a K-type thermocouple which is fixed 2 mm below the heater. In the present configuration, no provision is currently available for the measurement of surface temperature of the wafer. Therefore, the temperature measured by the thermocouple could be slightly different than the actual surface temperature. However, substrate temperature being one of the parameters to be optimized during MOVPE growth, a small difference in the two values will not affect the final outcome significantly. The substrate holder can carry wafers of 1" square size. The gap between the surface of wafer holder and showerhead is kept about 5 cm.

Safety is a key criteria for the design of an of MOVPE system. Note that the threshold limit value (TLV) of NH_3 is only 25 ppm while Hydrogen being a highly inflammable gas carries a low explosive limit (LEL) of only 4%. It therefore requires that the necessary safety issues are carefully considered. The first and foremost requirement is to ensure the leak integrity of overall MOVPE system which is better than $2x10^{-10}$ Torr Litre/sec in our case. All the gas handling components are properly enclosed in metallic cabinets and are provided adequate exhaust ventilation. It ensures that the gas cabinets are kept at low pressure with respect to the room under all conditions. Furthermore, online gas monitors for H_2, NH_3 and Silane are installed at key locations inside the MOVPE system. Suitable hardware interlocks to act against gas leak, chilled water failure, exhaust ventilation failure etc. are also incorporated in the control system.

3. Experimental detail

GaN epilayers are grown by using the indigenously developed MOVPE system on (001) Sapphire substrates at 70 mbar reactor pressure. Prior to the growth, Sapphire substrates are thoroughly cleaned with the help of organic solvents. Thereafter, the substrates are annealed under H_2 ambient at 1060 °C for 10 min. A nitridation step is then performed at the same temperature for 5-minute duration in presence of Ammonia with a flow of 1620 sccm. After this step, the reactor temperature is lowered in the range of 450 - 550 °C in order to grow LT GaN buffer layers. LT GaN is grown at a very slow rate typically in 30 mins duration with V/III ratio of 3178. Annealing of LT GaN buffer layer is thereafter performed in a temperature range of 950-1040 °C for duration varying from 30 sec. to 5 min. The optimized conditions are reached by analysing the crystalline quality of LT GaN layers by performing HRXRD measurements on a Panalytical X'Pert PRO MRD system. A hybrid monochromator (Goebel's mirror with a four-bounce crystal monochromator), which gives Cu $K_{\alpha 1}$ (wavelength = 1.54056Å) output with a beam divergence of ∼ 20 arcsecs, is used for HRXRD measurements. The optical quality of GaN layers is studied by performing PL measurements with 325 nm laser as the excitation source and a cooled charge coupled device as the detector. Surface morphology of MOVPE grown GaN epitaxial layers is studied by Field Emission Scanning Electron Microscopy (FESEM) technique. AFM measurements are performed using a multimode scanning probe microscope (NT-MDT, SOLVER-PRO, Russia).

4. Result and discussion

Note that the main objective of this article is to develop an indigenous MOVPE reactor which is affordable but can deliver the layers of desired quality. High quality GaN epitaxial layers have been routinely grown by numerous researchers worldwide during the last 3 decades [3, 24]. It has already established MOVPE as an excellent technique for this purpose. In this section, we illustrate the basic capabilities of our MOVPE system for the growth of GaN epitaxial layers.

4.1 *Optimization of GaN buffer layer*

Incorporation of LT GaN layer is a critical step in the epitaxial growth of thick GaN by MOVPE [24–28]. The mechanism by which it helps in the improvement of the crystalline quality of nitride epitaxial layers has been debated for a long time [26]. It is also argued that the minimum duration required for the annealing of GaN buffer layer is 6 min at 1125 °C [24]. However, the annealing duration must depend upon the thickness of buffer layer and the annealing temperature. After trying various complicated annealing procedures, it is concluded that the optimal annealing temperature is 850 °C [26]. However, there are reports which says that the optimum temperature for the growth of LT-GaN layer is 475 °C [27]. On the other hand, it is also reported that the ramp rate of annealing step is very critical for optimizing the quality of GaN epilayers where the growth temperature of LT-GaN was chosen to be 600 °C [28]. Similarly, an optimum thickness of LT-buffer is believed to exist [14], however, there are reports where good quality GaN epitaxial layers are grown without any buffer layer [29]. It therefore indicates that the optimization of LT-buffer layer is a very complicated process. Moreover, the growth parameters in a dynamic MOVPE process are interdependent. This makes the optimization process rather difficult. In view of this, a systematic process for the optimization of LT-GaN buffer is necessary and the same is discussed here. At first, the growth of un-coalesced low temperature GaN nucleation layer is performed on c-plane Sapphire substrate under the conditions listed in table 1. From a set of growth experiments (not shown here), it is observed that a reasonable layer quality of GaN buffer layer is obtained at a temperature of ∼550 °C. Hence, results from LT GaN layers grown at this temperature are only presented in this article. After the growth of LT GaN, an annealing step was completed and the LT GaN layers were taken out for the HRXRD measurements. HRXRD, being a non-destructive technique, is generally preferred for accessing the crystalline quality of grown layers by measuring the full width at half maximum (FWHM) of Bragg diffraction peaks.

It is known that the broadening i.e. the FWHM of HRXRD pattern is affected by defects and dislocations that might be present in the layer. Hence, a low value of FWHM is an indication of the good crystalline quality of epilayers. Several factors like the lateral coherence length (LCL), vertical coherence length, tilt and twist are usually associated with GaN epilayers grown on foreign substrates. Researchers often estimate the value of edge and screw dislocation density in GaN layer by measuring the value of FHWM of symmetric and asymmetric diffraction patterns respectively. Williamson-Hall analysis is performed on the symmetric (002), (004), (006) planes which are perpendicular to the growth directions (001) for estimating the value of microstructure. For this purpose, the experimental HRXRD data is first converted to the coordinates in reciprocal lattice units (q) using the following formulae [30, 31],

$$q_x = \left(\frac{1}{\lambda}\right)[\cos\omega - \cos(2\theta - \omega)] \qquad (1)$$

$$q_z = \left(\frac{1}{\lambda}\right)[\sin\omega + \sin(2\theta - \omega)] \qquad (2)$$

Here, λ is the wavelength of incident x-ray, ω is the angle at which the incident beam meets the sample surface, and 2θ is the angle at which the diffracted beam deviates from incident beam direction. FWHM of ω-scans in reciprocal lattice space for symmetric (002), (004) and (006) reflections obtained from the fitting of pseudo-Voigt line profile for all four samples are listed in table 1. As can be seen from table 1, the best crystalline quality of the sample is recorded in case of sample B4. Note that the quality of layer does not improve further by increasing either the annealing temperature or duration. A similar observation has been made in an earlier report [24]. HRXRD scans of sample B4 corresponding to symmetric (002), (004) and (006) reflections are shown in figure 3. The fitting of ω-scans by the pseudo-Voigt line profile is also shown by the red line in respective figures.

According to the conventional analysis, the fitted line shape profile accommodates two primary broadening mechanisms namely: finite lateral coherence length (LCL) and the tilt angle α. Among the two factors, LCL is independent of q whereas tilt contribution is known to be proportional to q. In case of pseudo-Voigt profile, the contribution of the two factors can be expressed as,

$$(\Delta q_{obs}(\omega))^n = \left(\Delta q_{(00l)LCL}\right)^n + (\alpha_{tilt} * q)^n \qquad (3)$$

where $\Delta q_{obs}(\omega)$is the FWHM of ω scan, $\Delta q_{(00l)LCL}$ and α_{tilt}define the contribution to the broadening due to finite LCL and tilt between sub-grains respectively. The values of $\Delta q_{obs}(\omega)$are obtained by fitting the diffraction pattern with pseudo-Voigt line profile. It represents the total broadening of ω-scan in q-space. The instrumental broadening (∼20 arcsec) is neglected since it is much lower than the FWHM of diffraction patterns. The factor n in the superscript is related to the fraction (f) of Lorentzian component in the pseudo-Voigt profile defined as follows [32],

$$n = 1 + (1 - f)^2 \qquad (4)$$

For perfect Lorentzian curve f is 1 and for perfect Gaussian curve f is zero. The Williamson-Hall plot for sample B4 using ω-scan in q-space for symmetric reflections is shown in figure 3(d). The value of n is determined from the pseudo-Voigt fitting using equations (1)–(4). The value of LCL and α is obtained from the straight line fit of equations 3. A similar procedure is carried out for all the other samples and the results are shown in table 2.

From table 2, it is clear that the structural quality of buffer layer B4 is superior among all the buffer layer sample having the maximum LCL and minimum tilt between the sub-grains.

Table 1. MOVPE Growth parameters of LT GaN buffer layer and Full width at half maxima (FWHM) of HRXRD pattern. The values of FWHM estimated from (002), (004) and (006) scans are given in reciprocal units (rlu) and degrees.

Sample No.	V/III ratio	Buffer growth temp. (°C)	Buffer growth time (min.)	Annealing temp. (°C)	Annealing time (s)	FWHM (002) rlu (deg)	FWHM (004) rlu (deg)	FWHM (006) rlu (deg)
B1	3178	550	30	950	30	0.0361 (5.42)	0.0641 (4.80)	0.0914 (4.54)
B2	3178	550	30	1000	30	0.0388 (5.76)	0.0675 (5.00)	0.1189 (4.71)
B3	3178	550	30	1040	30	0.0285 (4.26)	0.0519 (3.86)	0.0740 (3.67)
B4	3178	550	30	1040	300	0.0169 (2.53)	0.0322 (2.39)	0.0430 (2.13)

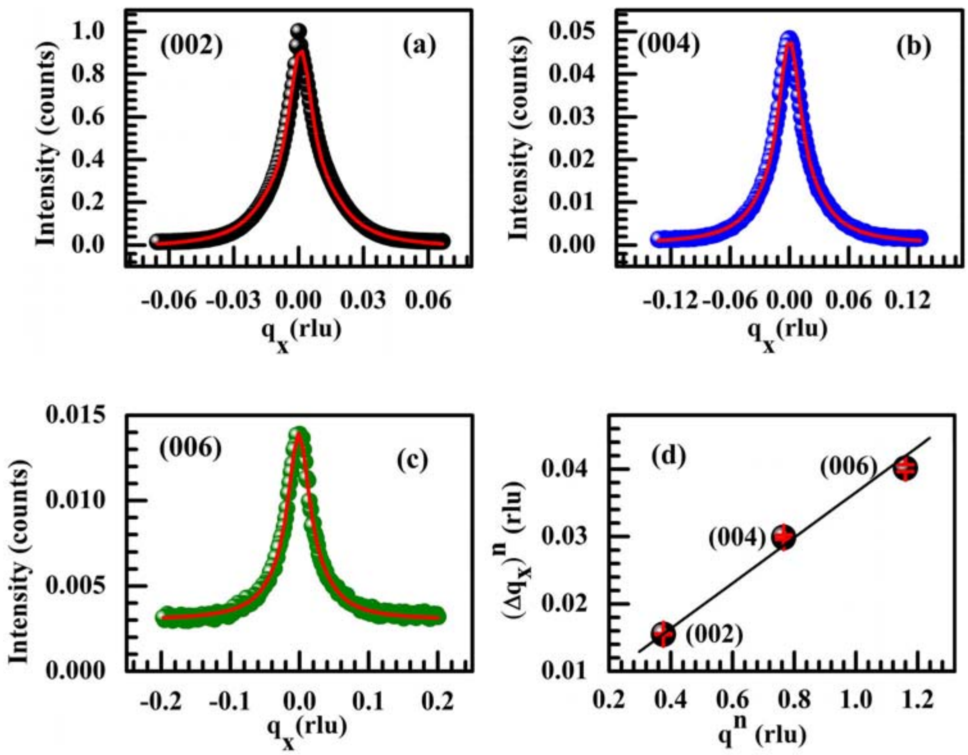

Figure 3. Intensity versus q_x curves for sample B4 (a) (002), (b) (004), and (c) (006) reflections. The pseudo-Voigt fitting curve is shown by the overlaying red line (d) Williamson-Hall plot for sample B4 by using (002), (004), and (006) reflections. The error bar shown in the data points represent the error contribution obtained from the pseudo-Voigt fitting.

Table 2. The values of *n*, LCL and tilt angle for all the LT GaN buffer layers obtained from Williamson-Hall analysis.

Sample no.	*n*	LCL (nm)	Tilt (degree)
B1	1.007	11.7	4.1
B2	1.07	12	3.5
B3	1.005	17.5	3.3
B4	1.02	20.2	1.8

4.2 *Optimization of the growth of GaN epitaxial layer*

After optimizing the growth of LT GaN buffer layer, next we discuss the influence of these parameters on the properties of high temperature bulk GaN (HT-GaN) epitaxial layers. Following the aforementioned results, the optimized values of annealing temperature and time are taken to be 1040 °C and 5 minutes respectively. Thickness of GaN buffer layer is varied by changing the growth duration from 2-12 min as shown in table 3. Other growth parameters for LT GaN layer are given in table 1. Growth temperature, V/III ratio and nominal thickness of HT-GaN layer were kept as 1020 °C, 4273 and 1 μm respectively. Thickness uniformity of GaN samples is tested by performing normal incidence reflectance measurements which is found to be ± 5% after 5 mm edge exclusion. Note that the measured value of thickness uniformity is achieved without incorporating substrate rotation, which is therefore reasonable.

In order to test the crystalline quality of GaN epitaxial layers, X-ray diffraction (XRD) patterns over broad angular range are recorded. Figure 4 shows the representative XRD pattern of sample T3. Diffraction peaks corresponding to GaN epitaxial layer are seen at 34.556° and 72.943°, which are labelled as (**002**) and (**004**) reflections of wurtzite GaN respectively. It indicates that the film is strongly c-axis oriented. In addition to the peaks corresponding to GaN, two more features are seen in figure 4 which are labelled as (**006**) and (**00.12**) diffraction peaks of sapphire substrate.

Epitaxial nature of thick GaN epilayers is confirmed by performing ϕ-scans where a representative pattern for sample T3 is shown in figure 5(a) for (**105**) skew-symmetric reflection of GaN. Observation of six distinct peaks separated by 60° clearly confirms the growth of wurtzite GaN epitaxial layer. In order to explore the epitaxial relation between the film and substrate, a similar ϕ-scan for ($\mathbf{10\bar{1}0}$) skew-symmetric reflection of Sapphire substrate is also recorded and the same is shown in figure 5(b). It is observed that the (**105**) plane of GaN is aligned with the

Table 3. Growth duration and thickness of LT GaN buffer used in the growth of thick GaN epilayer.

Sample No.	Buffer growth time (min.)	Buffer layer thickness (nm)
T1	12	48
T2	06	24
T3	04	16
T4	02	8

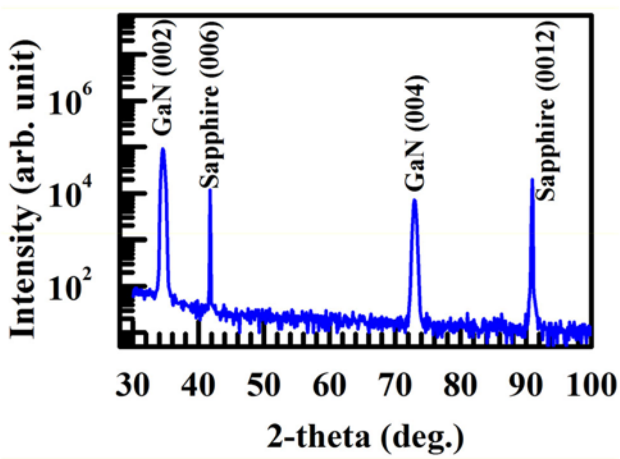

Figure 4. XRD pattern of sample T3 shown over a broad angular range.

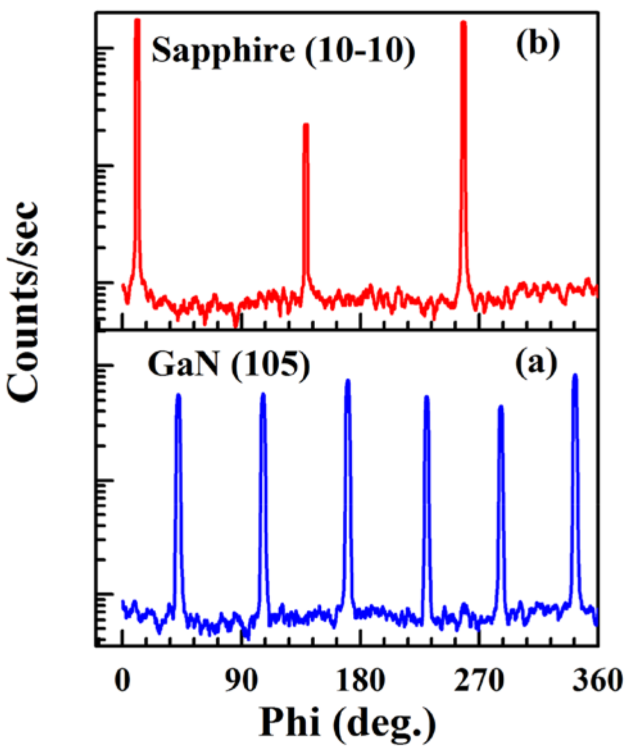

Figure 5. (a) Phi scan corresponding to (105) reflection of GaN epitaxial layer, (b) for $(\mathbf{10\bar{1}0})$ reflection of Sapphire for sample T3.

$(\mathbf{10\bar{1}0})$ plane of Sapphire, where the unit cell of GaN is rotated by 30° with respect to that of Sapphire.

Figure 6 shows the reciprocal space map of sample T3 for (**105**) reflection where sharp contours corresponding to GaN layer are observed. It clearly shows that the in-plane broadening of layer is larger than the out-of-plane broadening, which is usually observed in case of GaN epitaxial layers grown on foreign substrates [33].

Furthermore, a set of symmetric (002) and asymmetric (102) ω-scans for all the HT-GaN epitaxial layers are shown in figure 7. As can be seen from figure 7, the formation of defects/dislocations causes significant broadening in both the symmetric and asymmetric diffraction patterns for sample T1 where the values of FWHM for the two patterns are 1416 and 1550 arcsec, respectively. It is obvious from figure 7 that the crystalline quality of HT-GaN layer improves when the buffer layer deposition time is reduced from 12 to 4 min. However, a further reduction of the growth duration to 2 min. leads to a significant degradation of the crystalline quality of GaN layer in case of sample T4. In case of sample T3, the lowest value of FWHM of symmetric and asymmetric diffraction patterns is recorded. It clearly indicates a considerable reduction in both the edge and screw dislocation density. Moreover, the evident broadening of asymmetric reflection compared to the symmetric one indicates the dominance of pure edge dislocations in GaN epilayers. Note that the symmetric diffraction peak is broadened mainly by screw or mixed dislocations whereas edge dislocations are known to affect the broadening of asymmetric scans.

The measured values of FWHM of symmetric and asymmetric diffraction patterns of GaN epitaxial layer are comparable to those reported in literature. For example, a value of 506 arc second for 3 μm thick GaN epitaxial layers was reported recently which was shown to fall below 300 arc second at large annealing durations [24]. In that case, the growth temperature of thick GaN layers is about 100°C larger than the value chosen by us. Further, the thickness of

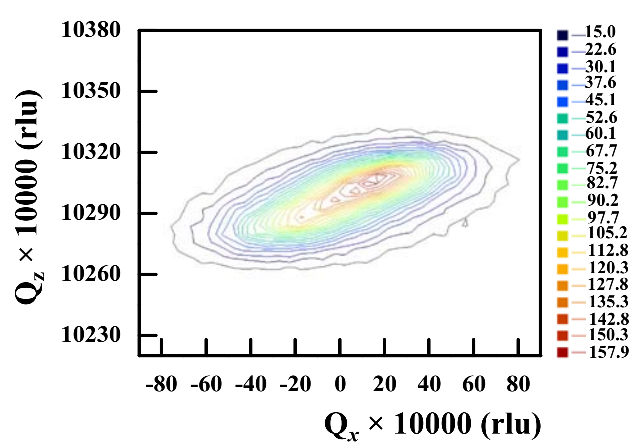

Figure 6. Reciprocal space map of sample T3 for (**105**) reflection.

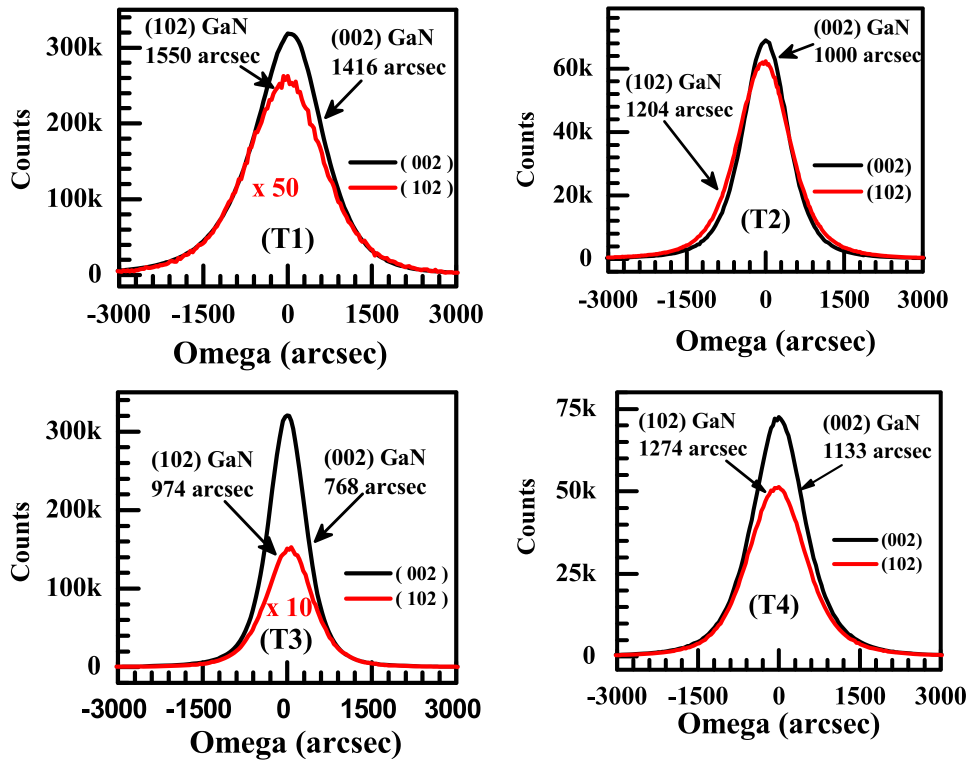

Figure 7. Symmetric (002) and asymmetric (102) HRXRD diffraction patterns for T1, T2, T3, and T4 samples respectively.

GaN epilayer in our case is only 1µm. It is known that the FWHM of (002) diffraction pattern decreases as a function of thickness of GaN epitaxial layers grown on Sapphire [34]. In past, similar or even higher values of FWHM are reported by other researchers for GaN epitaxial layers grown by MOVPE technique [3]. Very recently, even much larger values are reported for GaN epitxial layers that are grown by other techniques like pulse laser deposition and magnetron sputtering [35]. In view of this, the values of FWHM of diffraction pattern reported by us are highly reasonable. It confirms a good crystalline quality of GaN epitaxial layers grown by the indigenously developed nitride MOVPE system. From the values of FWHM for symmetric (002) and asymmetric (102) reflections, the dislocation density can be estimated as follows [36],

$$D_{screw} = \frac{\beta^2_{(002)}}{9b^2_{screw}}, D_{edge} = \frac{\beta^2_{(102)}}{9b^2_{edge}} \quad (5)$$

$$D_{dis} = D_{screw} + D_{edge} \quad (6)$$

Here, D_{screw} & D_{edge} are the screw and edge dislocation density, β stands for the FWHM of corresponding diffraction pattern, and b is the Berger vector length (b_{screw} = 0.5185 nm, b_{edge} = 0.3189 nm). The values of screw and edge dislocation density that are estimated from HRXRD measurements for all the GaN templates are extracted using equation (5) and are plotted in figure 8. The minimum value of dislocation density is found to be $\sim 4\text{x}10^9$ cm^{-2} which indicates about the reasonable crystalline quality of GaN epitaxial layers. Note that a high value of dislocation density typically of the order of 10^{10} cm^{-2} is usually reported for MOVPE grown GaN/Sapphire epitaxial layers [37, 38]. We ourselves have reported dislocation density of the same order in commercially available bulk GaN/Sapphire templates [38–40]. Such a large value of dislocation density originates due to the substantial lattice mismatch

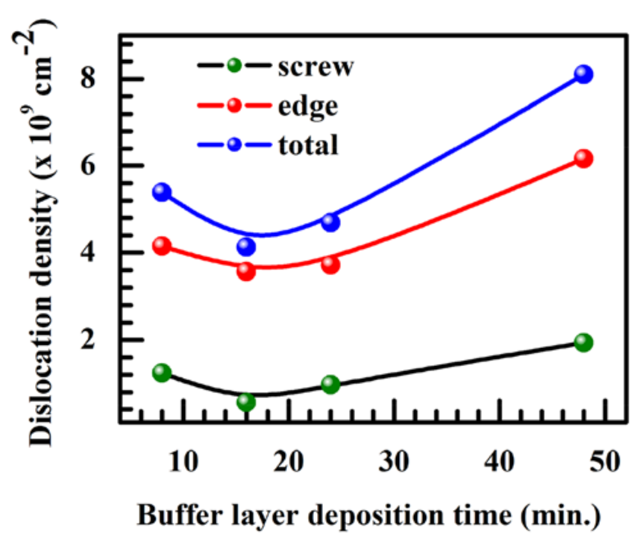

Figure 8. Edge, screw and total dislocation density for HT-GaN epilayer plotted as a function of buffer layer thickness.

between Sapphire and GaN. A large value of misfit strain causes the lattice of overgrown layer to be relaxed via the formation of defects/dislocations.

HRXRD and high resolution transmission electron microscopy (HRTEM) are the techniques preferred for the identification of the nature and distribution of dislocations. Surely, HRTEM is the most direct technique that furnish unambiguous evidences of dislocations present in the epilayer. However, the information obtained by this technique is of microscopic nature, which might not be a correct representation of the sample quality. Moreover, the technique also requires time-consuming sample preparation and is destructive. In view of this, HRXRD is usually favoured since the layer quality can be evaluated rather quickly by simply comparing the FWHM of the corresponding diffraction peaks of each samples, which helps the epitaxial growers in preparing the next growth recipe. In this method, all types of dislocations are generally considered to broaden the diffraction peaks and a low value of FWHM is believed to be a confirmation of good crystalline quality.

From the HRXRD results, it is possible to estimate the values of lattice constant in HT-GaN epitaxial layers. For this purpose, one needs to record the conventional ω-2θ scans for (002) and (102) reflections for GaN epilayer where representative scans for the samples T2, T3 and T4 are shown in figure 9. Since, the crystalline quality of T1 sample is very poor, it is therefore not included in further comparison. The lattice spacing corresponding to (002) and (102) crystallographic planes can be estimated by using the following method [32, 41]. For hexagonal system, the in-plane and out of plane lattice constants (a and c) are connected with inter-planar distance d_{hkl} for (h k l) planes by following relation:

$$\frac{1}{d_{hkl}^2} = \frac{4}{3}\frac{h^2 + k^2 + hk}{a^2} + \frac{l^2}{c^2} \tag{7}$$

Therefore, two ω/2θ scans for (002) and (102) reflections are recorded to find inter-planar spacing (d_{002} and d_{102}) from the peak positions $2\theta_{002}$ and $2\theta_{102}$ in the ω/2θ scans using Bragg's law. Then, the inter-planar spacing i.e. the lattice constants 'a' and 'c' are calculated by using equation (7) and are listed in table 4.

It is found that the value of in-plane lattice constant 'a' turns out to be smaller than the reported value of in-plane lattice constant of bulk GaN for all the samples. It indicates about the presence of a compressive strain in all three samples. In view of this, the values of strain in the perpendicular ($\varepsilon_\perp$) and parallel (ε_{II}) direction of GaN layers are also estimated by using the following relation [42],

$$\varepsilon_\perp = \frac{c_s - c_0}{c_0} \quad \text{and} \quad \varepsilon_{II} = \frac{a_s - a_0}{a_0} \tag{8}$$

where c_s (c_o) and a_s (a_o) are the lattice constants of strained (strain free) GaN layer in perpendicular (in-plane) direction respectively. For unstrained bulk single crystal, the value of a_o and c_o are taken as 3.1892 Å and 5.1850 Å respectively [43]. The in-plane biaxial stress σ_{xx} in GaN can be calculated as

$$\sigma_{xx} = \left(C_{11} + C_{12} - 2\frac{C_{13}^2}{C_{33}}\right)\varepsilon_{ll} \tag{9}$$

where C_{11}, C_{12}, C_{13} and C_{33} are the components of the elastic stiffness tensor of GaN and their values are taken from the literature [43]. The values of in-plane (a) and perpendicular (c) lattice constants, residual strain and stress are measured for GaN from HRXRD measurements and are listed in table 4.

From table 4, it can be seen that in all cases the GaN epilayer on Sapphire substrate is under compressive stress. It is reported in literature that GaN epitaxial layers grown on Sapphire are generally under compressive stress [43]. In our case, it is noted that the epitaxial layer with 4 min. growth of LT buffer layer is consisted of the minimum compressive stress of -0.76 GPa. It indicates that there exists an optimum buffer layer thickness for the growth of GaN epitaxial layer where the values of dislocation density and stress can be minimized under the respective growth conditions.

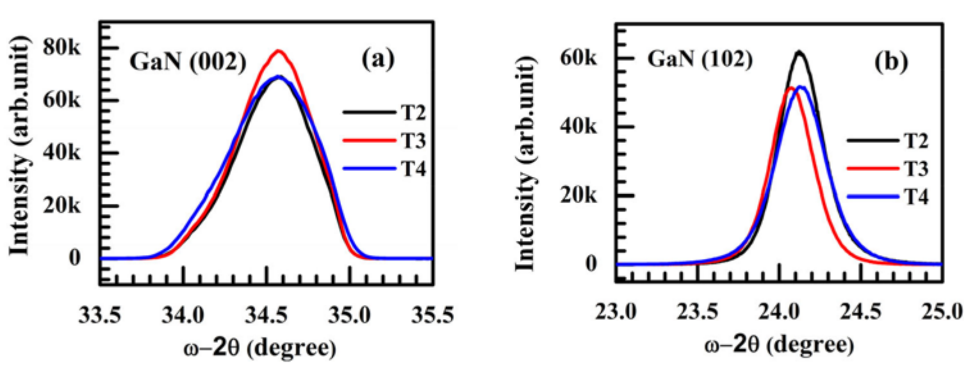

Figure 9. HRXRD of ω-2θ reflection of samples T2, T3 & T4 for (a) symmetric (002) and (b) asymmetric (102) planes.

Table 4. Lattice constants, strain and stress deduced from ω-2θ HRXRD scan for HT-GaN epilayer.

Sample No.	c-axis lattice constant (Å)	a-axis lattice constant (Å)	Strain ($\varepsilon_{\|}$)	Stress σ_{xx} (GPa)
T2	5.1874	3.1715	-5.53×10^{-3}	-2.48
T3	5.1871	3.1838	-1.69×10^{-3}	-0.76
T4	5.1887	3.1698	-6.07×10^{-3}	-2.73

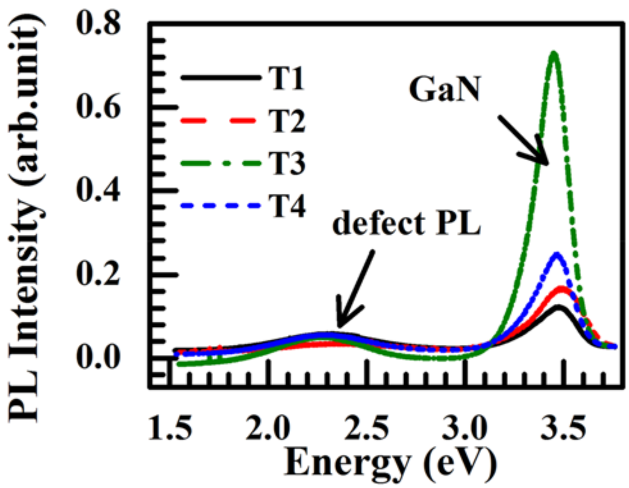

Figure 10. Room temperature PL spectra of GaN epitaxial layers.

Next, photoluminescence (PL) measurements can also provide useful information related to the optical quality of GaN epitaxial layers. PL measurements were performed on all the four samples and results are shown in figure 10. It is known that the residual stress in film modifies the band gap of a semiconductor. A compressive stress is known to increase the band gap of a semiconductor material. It is seen that the PL peak of all the samples appear at high energy when compared with the band gap of bulk GaN at room temperature [44]. Hence, all the grown layers are under compressive stress which is in good agreement with our HRXRD results. Furthermore, the PL peak of T3 sample appears at the lowest energy which confirms the observation made from HRXRD measurements where the particular sample is seen to possess the minimum compressive stress. PL intensity (FWHM) is measured to be the highest (lowest) in case of sample T3 respectively.

PL and HRXRD measurements therefore confirm that the crystalline quality of sample T3 is superior when compared with the remaining samples. Surface morphology of GaN epitaxial layers is studied by the SEM technique where representative images of the three samples are shown in figure 11. At large buffer layer thickness, an irregular surface morphology is recorded where a large density of islands/hillocks appear on the surface in case of sample T2 as obvious from figure 11 (a). A thick buffer layer leads to the initialization of 3-dimensional (3D) growth mode which may be the primary reason behind the uneven surface of sample T2. The surface morphology of grown layer improves significantly at low thickness of buffer layer as shown in figure 11 (b), where a mirror like surface is recorded. A good surface morphology of the particular sample is strongly corroborated by the results obtained from PL and HRXRD characetrization techniques. However, the surface morphology deteriorates when the buffer layer thickness is further reduced as shown in figure 11 (c). A very thin buffer layer leads to a partial coverage of Sapphire substrate which ultimately results in the porosity of GaN epitaxial layers. It is also supported by the trends seen in PL and HRXRD results.

A similar trend is also observed in the AFM images recorded on the same set of samples, where a representative picture for sample T3 is shown in figure 12. It is seen that the surface morphology of GaN layer is of reasonable quality where root-mean-square (RMS) roughness of film is measured to be ~ 3 nm. It should be noted that the films

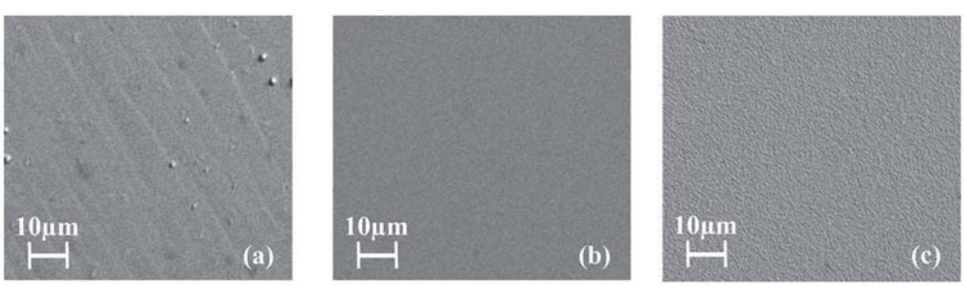

Figure 11. SEM images of GaN epitaxial layers grown by varying the buffer layer thickness, (a) T2, (b) T3 and (c) T4 samples.

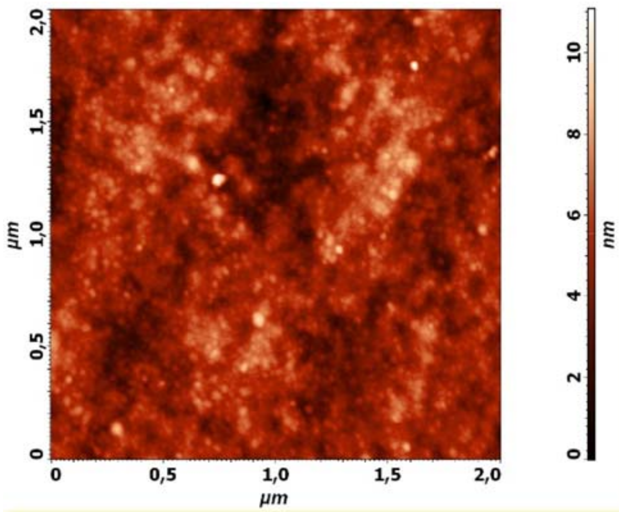

Figure 12. AFM image of sample T3.

are grown without any wafer rotation, which is known to improve the surface roughness along with the layer thickness uniformity drastically. In view of this, the measured value of surface roughness is reasonable. As mentioned earlier, the main aim of present article is to develop an indigenous MOVPE system which is capable of growing GaN epitaxial layers and the same is successfully demonstrated here. The quality of epitaxial layers will eventually improve with further optimizations and a few more improvisations in the MOVPE system. More efforts in this direction are in progress.

5. Conclusion

In conclusion, a nitride MOVPE system is indigenously designed and built with a motivation to develop a simple epitaxy tool which is though affordable but can deliver the layers of desired quality. A detailed study on the growth of GaN epitaxial layers by indigenously built MOVPE system is also presented. Crystalline quality of LT buffer GaN layer is first optimized by varying the growth temperature, annealing temperature and duration. Afterwards, the quality of HT GaN layer is also optimized by varying the thickness of LT GaN buffer layer where an optimum value of layer parameters is achieved. The crystalline quality of GaN epitaxial layers is found to be of reasonable quality where a moderate value of dislocation density is measured. Williamson-Hall analysis is used to determine the values of tilt and LCL of LT GaN layers. Results of HRXRD are found to be in good agreement with the PL data. The present study is important in view of the fact that the indigenously developed MOVPE system is found to produce GaN epilayers of reasonable crystalline quality. Commercially available MOVPE systems are really expensive and thus are out of reach of common researchers. Moreover, any further modification of such systems is not easy and is generally offered at a very high cost. The present work therefore can help in saving the financial resources which might enable an access to the high end research equipments for several researchers.

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