Growth of Nonpolar a-plane GaN Templates for HVPE Using MOVPE on r-plane Sapphire

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In order to establish the growth of nonpolar GaN templates for subsequent overgrowth via hydride vapor phase epitaxy (HVPE) or subsequent device epitaxy we studied the growth of a-plane oriented samples on r-plane sapphire via metal organic vapor phase epitaxy (MOVPE). The growth parameters like reactor pressure, growth temperature and V/III-ratio for the nucleation layer as well as for the GaN main layer grown on top were systematically investigated. A 2 step growth procedure and SiN interlayers were introduced for defect reduction yielding to improved photoluminescence and x-ray rocking curve measurements. This also resulted in reduced in-plane anisotropy and surface roughness values. Hence we achieved high quality nonpolar a-plane GaN layers suitable for subsequent processes.

1. Introduction

GaN-based devices like light emitting diodes (LEDs) are usually grown in c-direction, which is (0001) in crystal notation. Due to the polar character of the group-III nitrides in this particular direction and the biaxial strain induced in InGaN quantum wells having a certain lattice mismatch to the GaN barriers, huge piezoelectric fields develop. Hence the so called quantum confined Stark effect (QCSE) leads to a spatial separation of the wave functions of electrons and holes [1]. Optoelectronic devices therefore suffer from a reduced recombination probability, a red shift of the emission wavelength and a backshift to higher energies for higher drive current [2].

A way to overcome (or to reduce) these negative effects is to grow in nonpolar (or at least semipolar) direction. Nonpolar is every direction perpendicular to the c-axis like for example $[11\bar{2}0]$ (a-plane) and $[10\bar{1}0]$ (m-plane). Because of the lack of real bulk GaN the structures have to be grown either on foreign substrates like r-plane sapphire leading to a-plane GaN [3], or m-plane SiC [4] or $LiAlO_2$ [5] which results in m-plane GaN. However, such unusual growth directions typically lead to highly defective material. Alternatively, sliced pieces from hydride vapor phase epitaxial (HVPE) grown GaN can be used as substrates [6], where such defect formation is drastically reduced. However, only small areas of a few square millimeters of such substrates are nowadays available for exremely high prices. To cut a long story short the perfect substrate is still missing.

Therefore we are currently investigating the optimized growth of nonpolar / semipolar templates which can be used for subsequent device epitaxy. Such quasi substrates should be preferably grown by hydride vapor phase epitaxy (HVPE) to make use of its huge growth rates of several 10 to 100 µm/h. As the nucleation on foreign substrates is usually quite challenging and requires a wide range of optimization parameters, we have investigated the growth of nonpolar GaN templates for the subsequent HVPE process by the more flexible method of metal organic vapor phase epitaxy (MOVPE).

2. Experimental Procedure

All samples studied here are grown on 2 inch epi-ready r-plane sapphire wafers resulting in a GaN growth in $(11\bar{2}0)$ direction. For the MOVPE growth a commercial horizontal flow Aixtron AIX-200/4 RF-S reactor with the standard precursors trimethylgallium (TMGa), trimethylaluminum (TMAl) and high purity ammonia (NH_3) was used. As carrier gas we used Pd diffused hydrogen. The process temperature was controlled by a pyrometer at the backside of the rotation tray so that all temperatures given in the text are not the real temperatures but only the pyrometer read-out. Before starting growth the substrates were exposed to an in-situ desorption step at 1200°C for 10 min in hydrogen atmosphere [7].

First, we have systematically varied the growth parameters of the AlN nucleation layer (NL) while keeping the growth parameters of the main GaN layer the same as for standard c-plane growth (pressure of 150 hPa, temperature of 1120°C, V/III-ratio of about 2475, growth rate of approx. 2.4 µm/h and a thickness of 2.2 µm [7]). Then the growth parameters of the nonpolar GaN layer itself have been investigated. These studies led to the idea of a two step growth process and to the introduction of in situ SiN interlayers for defect reduction. Applying a constant flow of silane (SiH_4) and ammonia (NH_3), SiN_x is deposited and acts as a nanomask [8,9]. The surface is fractionally covered with SiN that influences the morphology of the overgrown layer resulting in a defect reduction. For this purpose the optimum deposition time and position of that SiN interlayer(s) have been evaluated.

For all these steps we used the crystal quality of the main GaN layer as the figure of merit. This was analysed by x-ray diffraction (XRD) rocking curve measurements (XRC) as well as low temperature (14 K) photoluminescence (PL) spectra. The latter enabling the qualification of typical defects in non-polar layers like basal plane stacking faults (BSFs) or prismatic stacking faults (PSFs) [10,11]. The surface quality was evaluated by scanning electron microscopy (SEM), optical phase contrast microscopy (OM) and atomic force microscopy (AFM).

3. Results and Discussion

3.1 MOVPE grown nucleation layers

Different NLs have been tested (i.e. AlN-NL, LT-GaN-NL), but only the AlN-NL yields to a $(11\bar{2}0)$–oriented surface. For this nucleation different growth temperatures ranging from 1010°C to 1200°C have been investigated. XRCs were recorded for both symmetrical $(11\bar{2}0)$–reflections along and perpendicular to the in-plane c-direction and for the asymmetrical $(10\bar{1}2)$ reflection (Fig. 1). The latter is known to be sensitive to edge and screw

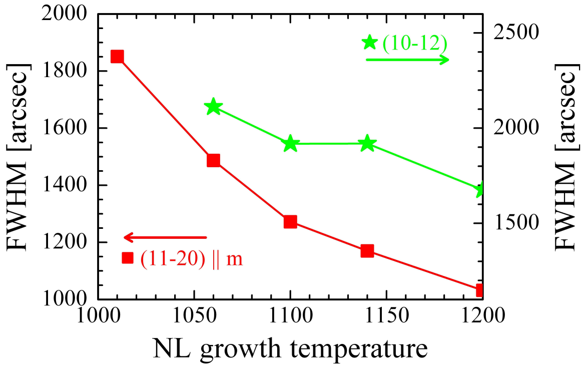

Fig. 1: FWHM of XRD rocking curve measurements of GaN layers with different temperatures during nucleation: $(11\bar{2}0)$–reflection along m-plane (squares, left axis) and $(10\bar{1}2)$–reflection (stars, right axis).

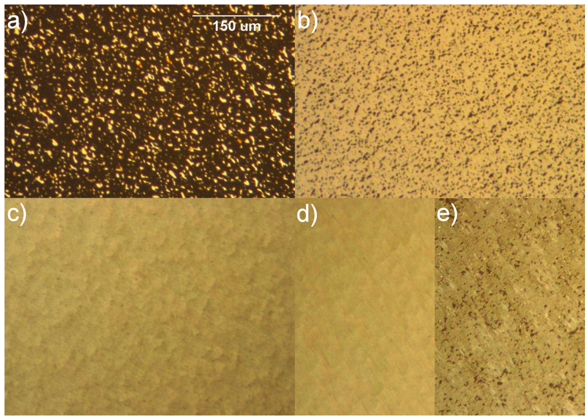

Fig. 2: Optical micrographs from GaN layers with different temperatures during nucleation: a) 1010°C, b) 1100°C, c) 1140°C, d) 1200°C (edge of wafer), e) 1200°C (center of wafer).

dislocations [12]. Obviously, the crystal quality increases with temperature as all FWHM values decrease. In parallel, the surface morphology improves (Fig. 2). At the lowest temperature of 1010°C, only a porous GaN layer was formed, while at 1100°C the layer is closed but suffers from lots of pits. For higher temperatures the pits decrease in number and size. R. Kröger reported that BSFs are generated at the interface to the nucleation layer [13]. It is also known that the crystal quality of AlN increases with increasing growth temperature. Therefore one can assume that the growth of good quality AlN as NL favors growth of good a-plane GaN. Unfortunately for too high temperatures the layers start to become inhomogeneous (see Fig. 2 d and e) probably due to wafer bending during growth. So the optimum temperature has been fixed at 1140°C.

Another challenge in AlN epitaxy (and therefore for this nucleation) are parasitic prereactions of TMAl and ammonia. They can be minimized by lowering the pressure, increasing the total flow and lowering the V/III-ratio resulting in a slight improvement of the surface morphology [14]. Moreover, the growth time of the NL has been varied between 5 min and 15 min without any obvious change in GaN quality. Hence, it has been set to 10 min, which results in a NL thickness of about 20 nm.

3.2 MOVPE grown GaN layers

After having optained reasonable conditions for the nucleation layer, the growth parameters of the main GaN layer were investigated. First, the *reactor pressure* was varied between 100 hPa and 200 hPa: At higher pressure, many pits develop on the surface (Fig. 3). By lowering the pressure, they can be reduced in size and density until they eventually vanish. In parallel, the PL intensity (Fig. 4) of the peak around 3.31 eV is decreasing with decreasing pressure. Paskov et al. [11,15] associated this peak to pyramidal stacking faults (PSFs) and partial dislocations (PDs). Hence lowering the reactor pressure reduces some structural defects. However for a too low pressure the XRD-FWHM values are increasing, while being constant on a fairly low level for higher pressure [16]. In parallel, the surface

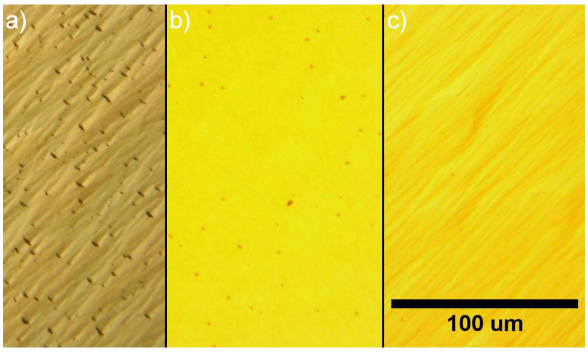

Fig. 3: Optical micrographs from GaN layers grown at different reactor pressure: a) 200 hPa, b) 150 hPa, c) 100 hPa. One can see the increased pit number and size with decreasing pressure.

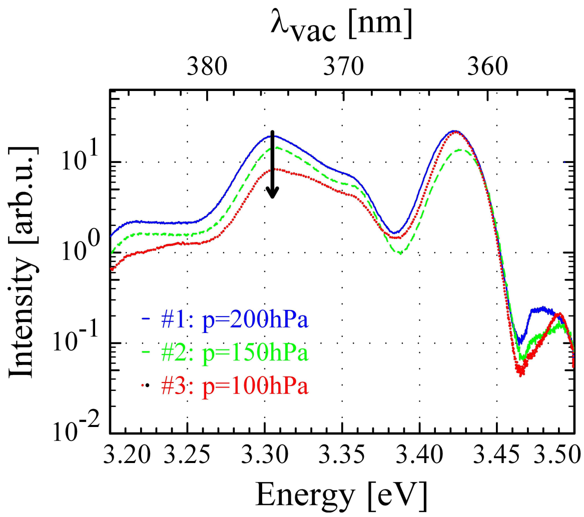

Fig. 4: Photoluminescence of GaN layers with different reactor pressure during growth. 100 hPa (dotted), 150 hPa (dashed), 200 hPa (solid).

gets roughened and develops a stripe like pattern at lower pressures. 150 hPa seems to be the best compromise between crystal quality and surface morphology.

The investigations on the *growth temperature* lead to an optimized temperature at about 1120°C. For lower values the widths of the XRCs increase as well as the PL intensity decreases. If the temperature is too high, the number and size of the pits increase as well as their luminescence [16].

Moreover, a series of samples has been grown with different *V/III-ratio*s ranging from 540 to 2180. The XRC measurements (Fig. 5) do not exhibit a clear trend on the first glance. We just notice a weak tendency to lower FWHM values of the asymmetrical reflections for increasing V/III ratio. In contrast to the crystal quality accessed by XRD, surface morphology is best for the lowest V/III-ratio (Fig. 6). This is confirmed by AFM measurements [16] where we found a root mean square (RMS) roughness of about 1.4 nm for a 5 µm × 5 µm scan of the respective sample.

3.3 2 step growth and introduction of SiN interlayers

The above described studies showed that for best bulk properties a high V/III-ratio and high temperatures are needed, whereas for best surface properties a low V/III-ratio and low pressure are favorable. Therefore we combined these two conditions by adopting a 2 step growth procedure. Best results were achieved for a variation of the V/III ratio only, from 2180 in the first layer with a thickness of approx. 1 µm to 540 in the second layer (thickness approx 1.2 µm) while holding constant the pressure at 150 hPa at a temperature of 1120°C. XRC FWHM values are plotted in Fig. 7 as dashed lines as reference for the following investigations. In order to further reduce the defect density, we studied in-situ

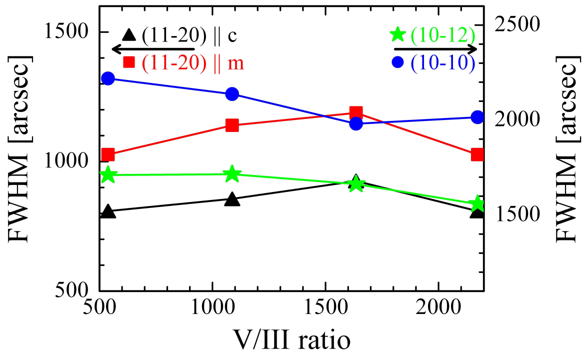

Fig. 5: FWHMs of XRC of GaN layers with different V/III ratio during growth. Symmetrical reflections are plotted on the left axis, asymmetrical ones on the right axis.

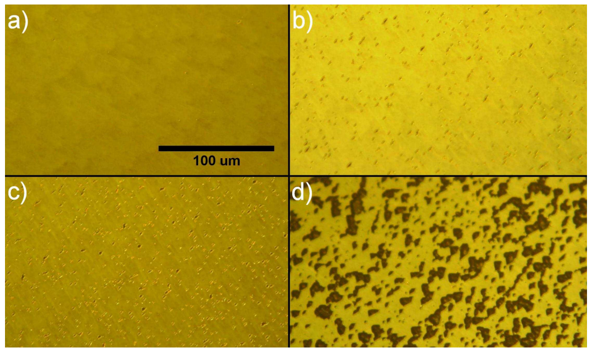

Fig. 6: Optical micrographs from GaN layers with different V/III-ratio during growth: a) 540, b) 1090, c) 1635, d) 2180. One can see the increased pit number and size with increasing flow of ammonia.

deposited SiN interlayers. We used standard conditions for SiN deposition as described elsewhere, showing that SiN can act as a defect reduction layer in c-plane oriented GaN [9]. When varying the SiN position within the GaN layer, we observed that a deposition directly on the nucleation does not help at all (Fig. 7). When shifting the interlayer towards the surface of the GaN, a fast decrease of the FWHM can be observed. Obviously, the blocking layer should be at least 150 nm above the NL with a weak optimum at about 300 nm. As most defects are created at the nucleation layer, we conclude that the SiN needs a certain distance to the defects to act as blocking layer.

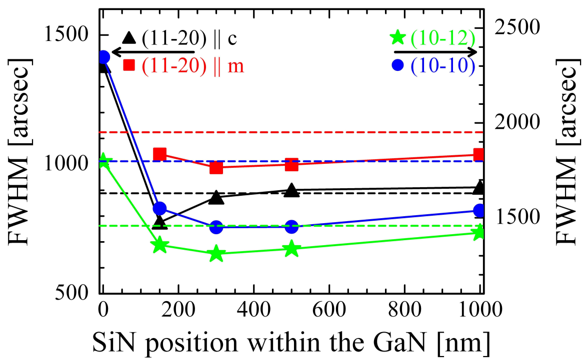

Fig. 7: FWHMs of XRC of GaN layers with introduced SiN interlayers plotted versus the SiN position within the GaN layer. The total thickness was kept constant at $\approx 2.2\,\mu m$.

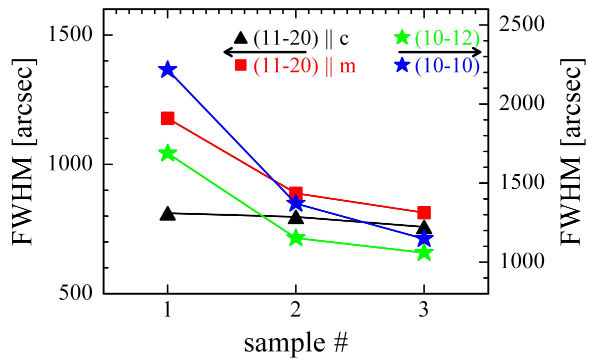

Fig. 8: FWHMs of XRC of a “simple” GaN layer (# 1) and samples with one (# 2) and two (# 3) SiN interlayers after $0.3\,\mu m$ and $0.3/1.0\,\mu m$, respectively.

Evaluations of the influence of the deposition time on the crystal quality were also made. By increasing the deposition time the XRC FWHMs decrease owing to a better crystal quality. However, at the same time the surface gets more and larger pits. By optimizing the growth parameters again and in particular increasing the thickness of the top GaN layer to about $2.3\,\mu m$, it was possible to achieve nearly pit-free surfaces.

Figure 8 shows a comparison of the XRC data from three different samples, without and with one and two SiN interlayers, respectively. The improvement in crystal quality is obvious, the FWHMs are drastically reduced down to values below 750 arcsec for the symmetrical $(11\bar{2}0)$ reflection and around 1050 arcsec and 1150 arcsec for the asymmetrical $(10\bar{1}2)$ and $(10\bar{1}0)$ reflections, respectively. The in-plane anisotropy, which seems to be undesirable for nonpolar growth [17] was reduced as well. This advancement in crystal quality was also confirmed by PL (Fig. 9). For GaN with one and two interlayers, not only the total intensity increases but also the near band edge emission (NBE) above 3.47 eV improves, while the luminescence from BSFs (≈ 3.42 eV, [11]) and other defects ($\approx 3.30 - 3.35$ eV, [11]) decreases relatively to the NBE. Additionally the surface roughness,

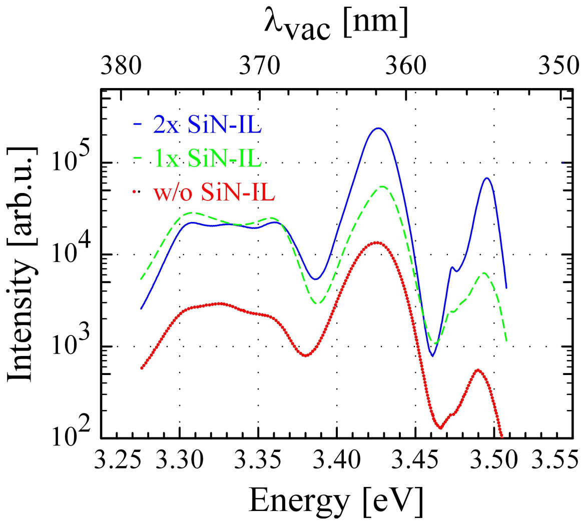

Fig. 9: PL of a "simple" GaN layer (dotted) and samples with one (dashed) and two (solid) SiN interlayers after 0.3 µm and 0.3/1.0 µm, respectively.

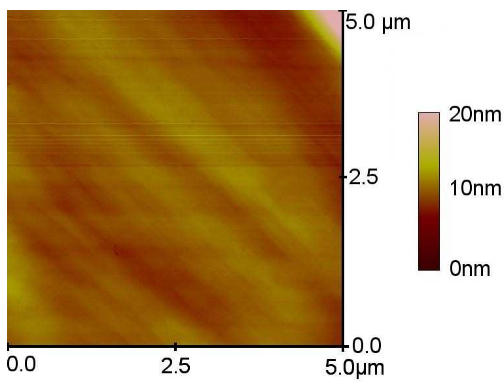

Fig. 10: 2D AFM measurement showing the smooth surface of an optimized a-plane GaN layer with two inserted SiN interlayers for defect reduction.

measured by AFM (Fig. 10) was reduced to values around 1.0 nm for a 5 µm × 5 µm scan.

4. Summary

By carefully optimizing the NL and the bulk GaN layer, we could achieve the growth of high quality nonpolar a-plane GaN. This was evidenced by small XRD FWHM values below 750 arcsec for the symmetrical $(11\bar{2}0)$ reflection and around 1050 and 1150 arcsec for the asymmetrical $(10\bar{1}2)$ and $(10\bar{1}0)$ reflections, respectively. Additionally, the PL of the NBE could be drastically improved by introducing a 2 step growth procedure and in particular by inserting SiN defect reducing interlayers. Furthermore, very smooth surfaces could be optained with an AFM RMS around 1.0 nm for a (5×5) µm^2 scan. In conclusion, we produced high quality nonpolar GaN templates suitable for subsequent overgrowth in HVPE.

5. Acknowledgments

This work was financially supported by the Deutsche Forschungsgemeinschaft within the research group "Polarization Field Control in Nitride Light Emitters (PolarCoN)" under contract no. Scho 393/22–1. The sample characterization by Wenshan Pang is gratefully acknowledged.

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