Improvement of MOVPE Grown $(11\bar{2}2)$ Oriented GaN on Pre-Structured Sapphire Substrates Using a SiN_x Interlayer and HVPE Overgrowth

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In this article two methods for improvements of $(11\bar{2}2)$ *oriented semipolar GaN grown by MOVPE on pre-structured sapphire substrates are investigated. The integration of a* SiN_x *interlayer helps to obtain a better crystal quality. Also the overgrowth of the MOVPE samples by HVPE is a way to obtain a smoother GaN surface. A high incorporation of oxygen on* $(11\bar{2}2)$ *oriented GaN compared to* (0001) *oriented GaN grown by HVPE was observed.*

1. Introduction

Despite longtime research on solid state lighting, there are still a lot of challenges for highly efficient, bright LEDs in the green region of the optical spectrum. In literature, this problem is referred to as the green gap. The high indium concentrations that are required for long-wavelength emission of GaN based devices give rise to strain in the piezoelectric material. This is caused by the significant lattice mismatch between GaN and the active GaInN quantum wells. This results in defects that increase the nonradiative recombination rate and high internal electric fields that lead to the so-called quantum-confined Stark effect (QCSE). Using semipolar crystal planes with reduced internal polarization fields, the impact of the QCSE might be reduced [1–5].

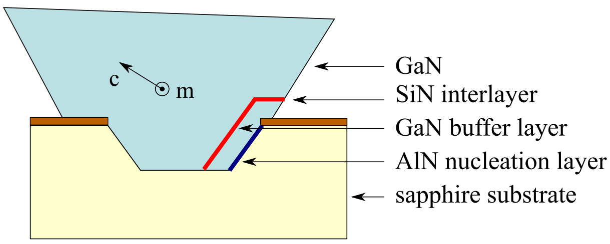

Fig. 1: Schematic structure of a single GaN stripe nucleating on the c-plane-like side facet of the etched trench. Structures grown on r-plane pre-structured sapphire wafers eventually coalesce to a closed layer with a $(11\bar{2}2)$ semipolar surface.

This article [6] concentrates on our studies of semipolar $(11\bar{2}2)$ oriented GaN grown by MOVPE on r-plane $(10\bar{1}2)$ pre-structured sapphire substrates (r-PSS). The growth of a semipolar $(11\bar{2}2)$ GaN layer on the sidewall of an r-PSS was first demonstrated by Okada et al. [7]. The investigated approach is sketched in Fig. 1: At first, a SiO_2 mask is deposited for selective epitaxy. Trenches are etched along a-direction into the sapphire substrates. One facet of the trenches is c-plane like. On these facets, GaN stripes grow in c-direction and finally coalesce to a closed semipolar layer.

A known method for the reduction of defects in conventional c-plane GaN is the in-situ integration of SiN_x interlayers [8–12]. This method is a special case of epitaxial lateral overgrowth (ELOG) [13] and has been studied extensively by our group on c-oriented GaN [14, 15] and AlGaN layers [16, 17]. It has been adopted successfully also to the structures described above [18]. Further investigations are presented in this article.

In another experiment, the MOVPE samples have been overgrown by hydride vapor phase epitaxy (HVPE).

2. Template Preparation and MOVPE Growth Conditions

The r-plane sapphire substrates are structured as described by S. Schwaiger [19]. At first the growth mask, a 200 nm thick SiO_2 layer, is deposited with PECVD on the bare r-plane sapphire wafer. By conventional photolithography, resist stripes with a period of 6 µm and a width of 3 µm are manufactured and an etch mask containing Ni is deposited. After lift-off, the trenches are etched by RIE using the gases Ar, BCl_3 and Cl_2. The remains of the metal mask are removed wet chemically.

GaN growth by MOVPE is carried out in a commercial Aixtron-200/4 RF-S HT reactor with the precursors TMGa, NH_3 and TMAl. Growth starts with the deposition of an AlN:O nucleation layer. A GaN buffer layer follows at 1105°C and a V/III ratio of 650. After 10 min, at a distance of approximately 1.2 µm from the c-facet, GaN growth is paused for the deposition of an in-situ SiN_x interlayer that is formed with the precursor SiH_4. The main GaN layer grows at 1025°C for 110 minutes at a V/III ratio of 647.

3. SiN_x Interlayer

In order to investigate the influence of different parameters for SiN_x deposition on the final GaN layer quality, the following experiments have been conducted:

At first, one sample without SiN_x interlayer and three samples with SiN_x interlayer, deposited at 1005°C, 1025°C and 1045°C, for 3 min, at a molar SiH_4 flux of 0.1 µmol min^{-1}, were produced. For all samples, the GaN growth temperature was unchanged. It is assumed that these conditions result in a submonolayer thickness of SiN_x.

In a next step, we have studied various thicknesses of the SiN_x interlayer by varying the deposition time between 2 and 7 minutes at a deposition temperature of 1025°C.

To eliminate the influence of variations of the sapphire templates that are caused by variations at processing, this study was performed on quarters of two 2" wafers. Here, samples from the same wafer are labeled series S1 and series S2, respectively.

Finally the influence of the position of the SiN_x interlayer was surveyed by varying the growth time of the subjacent GaN buffer layer. On one quarter of the used sapphire wafer, the SiN_x was deposited directly on the nucleation layer. Then the SiN_x interlayer was deposited after 5 min, 7.5 min and, as a reference, again after 10 min. Here, a SiN_x deposition time of 5 min at 1025°C was chosen.

3.1 Results

To compare the crystal quality, the full width at half maximum (FWHM) of high-resolution X-ray diffraction rocking curves (HRXRD RCs) was evaluated (Fig. 2 (left)). The smallest FWHM of this series was found for the symmetric ($11\bar{2}2$) reflection, measured parallel to the sapphire trenches, at a deposition temperature of 1005 °C. However, its ($11\bar{2}4$) rocking curve is the broadest of all. If the SiN_x interlayer is deposited at 1025°C, a clear improvement is visible compared to the sample without SiN_x. For higher temperatures an increase of the FWHMs is measured. Low temperature PL spectra (Fig. 5 (left)) show a slight decrease of the intensity of the peak related to basal plane stacking faults (BSF) at 3.42 eV for the SiN_x interlayer deposited at 1025°C. The curves have been normalized with respect to the peak of the donor bound exciton (D^0X) at 3.485 eV. Out-of-plane X-ray diffraction measurements performed at the synchrotron ANKA at Karlsruhe Institute of Technology revealed that similar samples of our group have an excellent stacking fault density with values down to $4.4 \cdot 10^3\,\mathrm{cm}^{-1}$ [20]. We assume that the stacking fault density of the samples investigated in this article is equal or even better. The results of atomic force microscopy (AFM) measurements show that the sample without SiN_x has the lowest roughness (Fig. 2 (right)). Its root mean square (RMS) value is 42 nm on an area of $50 \times 50\,\mu\mathrm{m}^2$. The sample with a deposition temperature of 1025°C has a comparable or only slightly increased RMS value of 53 nm. For higher and lower SiN_x deposition temperatures the surface quality is heavily reduced.

In Fig. 3, the results of the SiN_x deposition time series are depicted. For the FWHMs of HRXRD rocking curves, a minimum can be found at $t = 5$ min. There is no significant change in the surface roughness between 2 and 5 min, but the surface degrades heavily for longer deposition times. Regarding the PL spectra (Fig. 5 (right)), there is no clear relation for the BSF related peak to the deposition time for series S1. However, for series S2 a distinct increase of the BSF related peak with the deposition time is observed.

As can be seen in Fig. 4 a), decreasing the GaN buffer layer thickness leads to an improvement of the FWHM of the ($11\bar{2}2$) reflection. While the ($11\bar{2}4$) reflections seems to be unaffected, the here also investigated (0006) reflection has a minimum at 7.5 min and increases again for 5 min. There is no clear change in the AFM RMS value between 5 min and 10 min (Fig. 4 b)). Direct deposition on the nucleation layer leads to both broad RCs and a rough surface.

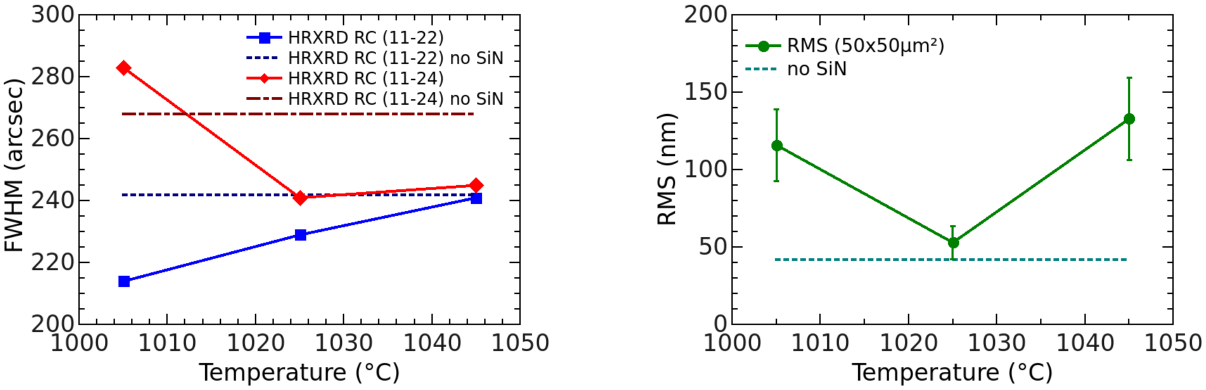

Fig. 2: HRXRD and AFM results of samples with SiN_x interlayer deposited at different temperatures compared to the sample without SiN_x.

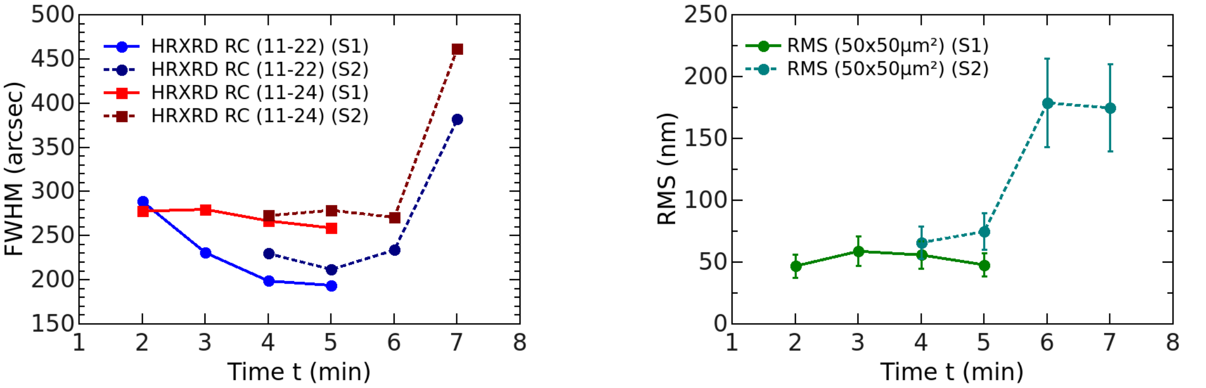

Fig. 3: HRXRD and AFM curves of samples with different deposition time of the SiN_x interlayer. Each series S1 and S2 is from one wafer that has been quartered before epitaxy, respectively.

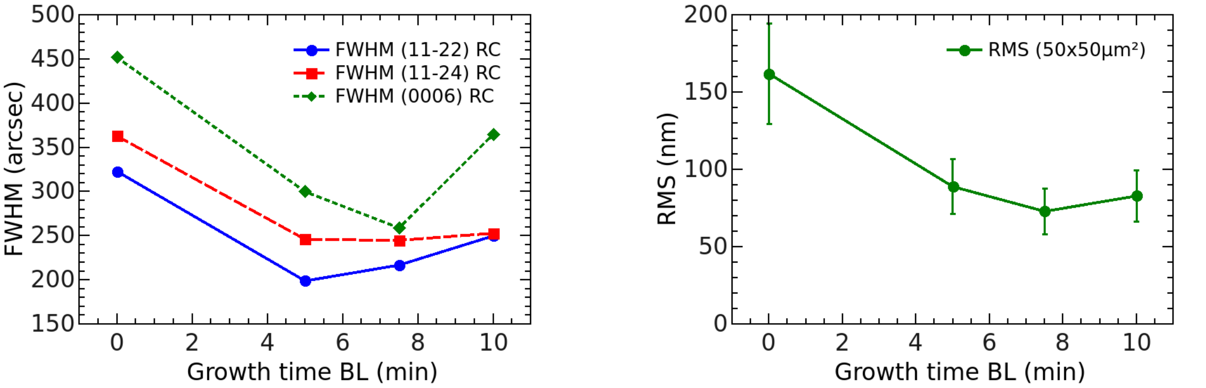

Fig. 4: HRXRD and AFM curves of samples with different positions of the SiN_x interlayer. The time indicates the growth time of the GaN buffer layer below.

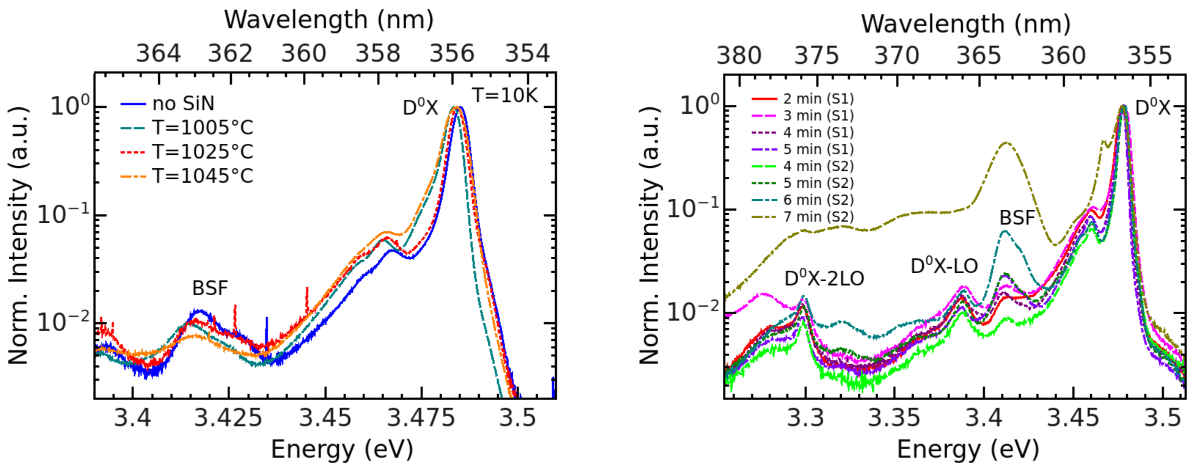

Fig. 5: PL spectra of samples with SiN_x interlayer deposited at different temperatures (left) and different time (right).

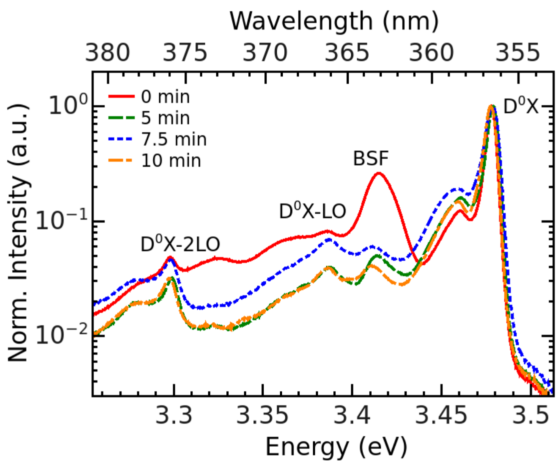

Fig. 6: PL spectra of samples with different positions of the SiN_x interlayer. The time indicates the growth time of the GaN buffer layer below.

Regarding the intensity of the BSF related peak in the PL spectra (Fig. 6), the sample with the interlayer deposited after 10 min is the best. The samples with 5 min and 7.5 min of buffer layer growth seem to have a slightly higher BSF density. Accordant to the HRXRD and AFM results, the sample with SiN_x deposited directly on the nucleation layer leads to the lowest crystal quality. In total, a growth time of 5 min of the GaN buffer layers seems to be the optimum.The AFM image of this sample is shown in Fig. 7.

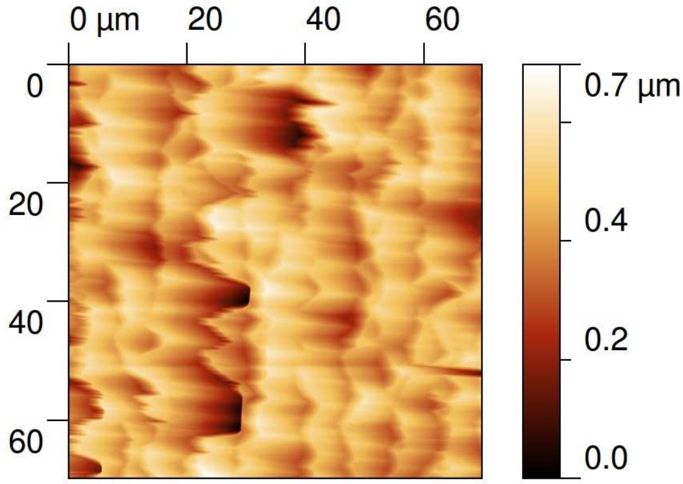

Fig. 7: AFM image of MOVPE grown sample with SiN_x interlayer deposited at 1025 °C, for 5 min after 5 min of buffer layer growth. The individual stripes have coalesced to a closed layer.

4. HVPE Overgrowth

Subsequently, we have overgrown a $(11\bar{2}2)$ MOVPE sample by hydride vapor phase epitaxy (HVPE) at our optimized c-plane conditions at a temperature of $T = 1069$°C. The growth starts at a V/III ratio of 1150 for 30 s. The main HVPE layer is deposited at a V/III ratio of 77 for 30 min. Then, for the last 10 min, the V/III ratio is increased to 230. At the end the V/III ratio is increased to promote 2D growth on a microscopic scale. The thickness of the obtained HVPE layer was approximately 46 µm. The FWHM of HRXRD rocking curves are 215" for the $(11\bar{2}2)$ and 236" for the $(11\bar{2}2)$ reflection, respectively. The surface roughness, measured by AFM (Fig. 9), dropped considerably to 14 nm on a measured area of $50 \times 50\,\mu m^2$.

By van der Pauw Hall experiments, a high carrier density of $n \approx 3 \cdot 10^{19}\,cm^{-3}$ was measured in such HVPE layers, whereas c-oriented samples grown under identical conditions show a carrier density of only $n \approx 2 \cdot 10^{16}\,cm^{-3}$. Secondary ion mass spectrometry (SIMS), performed on the $(11\bar{2}2)$ oriented sample, reveals a high oxygen concentration of $1 \cdot 10^{19}\,cm^{-3}$ similar as observed by M. Amilusik et al. on other semipolar planes [22] hence confirming the results of the van der Pauw measurement. In Fig. 8 the normalized PL spectra of the two HVPE grown samples are depicted. The high-energy shoulder at the $(11\bar{2}2)$ sample is the result of the high carrier concentration, and is caused by the free electron recombination band (FERB) [23].

5. Conclusions

The studies presented here have shown that the integration of an in-situ deposited SiN_x interlayer helps to improve the crystal quality of our semipolar $(11\bar{2}2)$ GaN layers deposited on patterned sapphire substrates on the possible cost of a slight increase of the surface roughness. PL measurements reveal a small decrease of the basal plane stacking fault density. We obtained best results at a deposition temperature of 1025°C and a deposition time of 5 min after 5 min growth time of the GaN buffer layer.

Overgrowing the MOVPE samples by HVPE results in a heavy reduction of the surface roughness. Compared to c-plane GaN, ($11\bar{2}2$) oriented samples show a favored incorporation of oxygen during growth that results in a high n-carrier concentration and a considerable signal above 3.5 eV in PL measurements that is induced by the free electron recombination band.

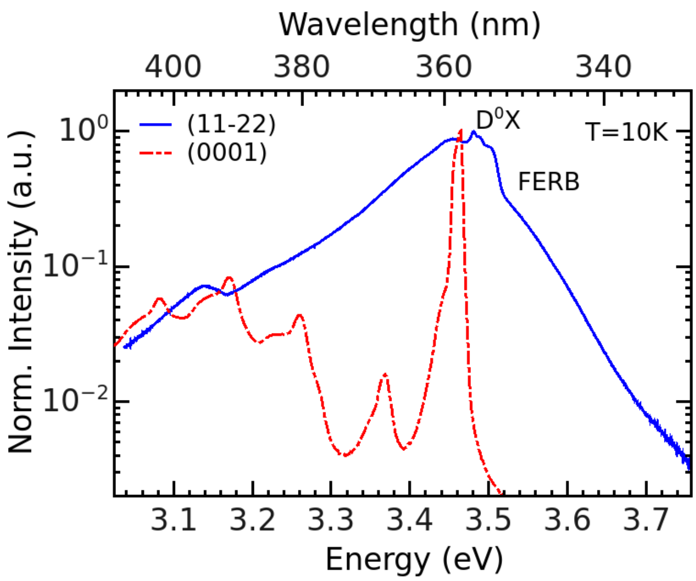

Fig. 8: Normalized low temperature PL spectra of a ($11\bar{2}2$) and a (0001) oriented HVPE grown GaN sample.

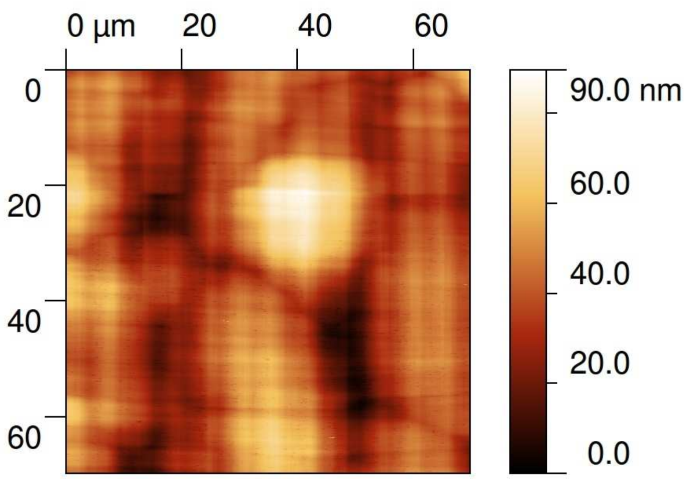

Fig. 9: AFM image of HVPE grown sample. The surface is much smoother and more homogenous compared to the MOVPE grown sample.

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