HVPE Growth on MOVPE-Grown Semipolar $(11\overline{2}2)$ GaN

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In this article HVPE layers deposited on MOVPE-grown (11 $\overline{2}2$) GaN are investigated. The properties of the MOVPE templates that are used for the HVPE experiments are described. HVPE overgrowth leads to a smoother surface and shows indications of a better crystal quality. For further improvements a HVPE growth temperature series was conducted.

1. Introduction

Despite of long-lasting research and development, there are still challenges in the production of light-emitting diodes (LEDs) for general lighting and other applications. While excellent LEDs in the red and blue spectral range are commercially available yet, there is still a lack of highly efficient LEDs in the green to yellow range. In literature, this problem is referred to as the green gap.

For the GaN material system, one reason for the breaking down of the efficiency is the socalled quantum confined Stark effect (QCSE). Since today's LEDs are grown in the common [0001] (c-)direction, the internal polarization fields are perpendicular to the quantum wells. The cause of these internal fields are spontaneous and piezoelectric polarization. The reason for the latter is mechanical strain that increases with higher In incorporation in the quantum wells, which is necessary for long-wavelength emission. This leads to a local separation of electrons and holes in the quantum wells. Thus their lifetime increases and the radiative recombination rate decreases. This is discussed to be a major reason for the decrease of the device efficiency [1-3].

One way out could be the use of crystal planes with reduced polarization fields. Crystal planes perpendicular to the (0001) plane are called nonpolar, while all other planes in between are named semipolar.

High indium incorporation in nonpolar planes turned out to be very difficult, but is mandatory for long-wavelength emission. Also those planes contain high defect densities which give rise to nonradiative recombination. Thus the growth of semipolar crystal planes is favored [2, 4].

Today homoepitaxy is not an option because of deficient availability and high cost of semipolar bulk GaN wafers. Here, we focus on a heteroepitaxial approach, described in the dissertation of S. Schwaiger [4], to produce semipolar $(11\overline{2}2)$ GaN:

As shown in Fig. 1, $(10\overline{1}2)$ r-plane sapphire serves as a substrate. Trenches with c-planelike side facets are etched into the substrate in a-direction of the sapphire. After masking



Fig. 1: Schematic of the production of $(11\overline{2}2)$ GaN substrates. During selective epitaxy, GaN stripes grow on the c-plane-like side walls of the trenches that have been etched into the sapphire substrate. After some time the individual stripes coalesce to a closed layer.

areas with SiO₂, selective growth with metalorganic vapour phase epitaxy (MOVPE) follows on the side facets of the trenches in the common c-direction with the following orientation: $c_{sapphire}||c_{GaN}$ and $a_{sapphire}||m_{GaN}$. r-plane sapphire is used because the angle of 57.61° between the (1012) sapphire surface and the c-direction is very close to the angle of 58.41° between the (1122) plane and the c-direction of GaN.

Eventually the single GaN stripes coalesce to a closed layer with (1122) surface. Subsequently, we have overgrown these templates by hydride vapour phase epitaxy (HVPE). This method provides substantially higher growth rates. Hence thick layers can be grown, which should help to improve the crystal quality. There is also the possibility to produce freestanding GaN substrates by this method. The feasibility has been shown by Yamane et al. [7].

2. Experimental

The structuring of the r-plane sapphire substrates as well as the MOVPE growth are described in detail in T. Meisch's contribution to this Annual Report (p. 52 ff., Sect. 2.). For the templates used in the studies described below, the GaN buffer layer is grown at 1100°C. Both the SiN interlayer and the low-temperature top layer grown at 1000°C are included.

Properties of the MOVPE templates A template that has been grown under the above described conditions shows full widths half maxima (FWHMs) of high-resolution X-ray diffraction rocking curves (HRXRD RCs) of 265" and 430" for the symmetric (11 $\overline{2}2$) reflection parallel and perpendicular to the c-direction, respectively and 337" for the asymmetric (11 $\overline{2}4$) reflection. Atomic force microscopy (AFM) measurements show a surface roughness of 82 nm (root mean square (RMS)) on an area of 20 × 20 µm².

2.1 HVPE overgrowth

For further improvements the samples have been overgrown by HVPE with the following conditions: growth temperature $T = 890^{\circ}$ C, pressure p = 900 hPa, V/III ratio = 17.5 and growth time t = 60 min. This experiment was done along the parameters described in [4]. In Sect. 2.2 they have been varied for further improvements.

HVPE results and comparison to the MOVPE template Scanning electron microscope (SEM) investigations of the cross-section (Fig. 2) show a clear material contrast between the MOVPE and the HVPE layer. The growth rate under the above described conditions is $12.3 \,\mu$ m/h.

The FWHM of the HRXRD rocking curve of the symmetric $(11\overline{2}2)$ reflections (parallel and perpendicular to the c-direction) dropped to 185" and 264", respectively, though the FWHM of the asymmetric $(11\overline{2}4)$ reflection increased to 365".

The AFM results are depicted in Fig. 3. The MOVPE template shows a sawtooth-like surface. The origin is the coalescence of the single GaN stripes and the misalignment of the c-directions of sapphire and GaN. After HVPE overgrowth, the surface turned into a bubble-like structure with reduced roughness of 42 nm (RMS) on an area of $20 \times 20 \text{ µm}^2$.

2.2 HVPE temperature series

In order to achieve a higher growth rate and to investigate the influence of the growth temperature, the experiment has been repeated with increased temperatures of 930° C and 970° C.

Direct results As shown in Fig. 4, the growth rate could only be increased a little bit at 930°C. At 970°C the growth rate drops again. The higher growth rate seems to influence



Fig. 2: SEM in-lens image of the HVPE sample. A clear material contrast between the HVPE and MOVPE layer is visible.



Fig. 3: AFM measurement of the MOVPE (left) and HVPE sample (right). With HVPE the sawtooth-like surface of the MOVPE sample became a bubble-like structure with reduced roughness.



Fig. 4: HVPE growth rate in [1122] direction (left) and FWHM of HRXRD RCs over temperature (right).

the crystal quality in a negative way. The FWHMs of the HRXRD RCs of the $(11\bar{2}2)$ reflections both increase. Interestingly the FWHM of the $(11\bar{2}4)$ reflection is decreasing, so no clear conclusion can be drawn out of this data. Also the normal process variation has to be considered.

The AFM measurements show more clear results (Fig. 5). The surface roughness decreases with higher temperatures. For a fair comparison, the area used to determine the RMS value has been increased to $50 \times 50 \,\mu\text{m}^2$.

Hillocks On all HVPE samples, huge hillocks emerged (Fig. 6). They are randomly distributed on the wafer with a slightly higher density at the wafer edge. The hillocks have a height of approximately $20 \,\mu\text{m}$, length of $145 \,\mu\text{m}$ and width of $75 \,\mu\text{m}$. They are smoothing iron shaped and are all oriented in the same direction perpendicular to the trenches in the sapphire in +c-direction. An electron backscatter diffraction (EBSD)

measurement revealed that they are grown monocrystallinely on the GaN layer. The origin is unknown at present and will be further investigated.



Fig. 5: AFM measurement of the samples grown at 890°C, 930°C and 970°C. Bottom right: the surface roughness decreases with higher temperature.



Fig. 6: Typical appearance of the hillocks observed on all HVPE samples. Left: SEM, right: optical microscope.

PL and **CL** measurements In Fig. 7 (left), the photoluminescence (PL) spectra of the HVPE samples are depicted. As a reference the PL spectrum of the MOVPE template has been inserted in this graph, too. It was necessary to scale it down by a factor of approximately 100, since it has a much higher intensity and was measured with another sprectrometer. The peak at approximately 3.48 eV can be assigned to the donor-bound exciton (D⁰X) transition and the peak at 3.46 eV might be related to acceptor-bound excitons (A⁰X). The reason for the peak at approximately 3.42 eV might be basal plane stacking faults (BSF). We observed a slight shift of the D⁰X peak energy, which is probably due to strain differences caused by varying growth temperatures and layer thicknesses. The peaks of the HVPE samples at lower energies are not as distinct as the peaks of the MOVPE sample. Also a high-energy peak above 3.5 eV was observed on all HVPE samples. The MOVPE sample does not show this peak. It is important to mention that the energy of the maximum shifts with higher growth temperatures to higher energies and the peak gets broader.

To investigate its origin, cathodoluminescence (CL) measurements of the cross-section have been performed. Also here a clear material contrast between the HVPE layer and the MOVPE layer was detected (Fig. 8). Local CL spectra show the high energy-peak that was measured with PL, too. As can be seen in Fig. 7 (right), it clearly belongs to the HVPE layer.

To ensure that no Al content is causing this peak, a secondary ion mass spectrometry (SIMS) measurement was done. The Al concentration was below the detection limit.



Fig. 7: Left: PL spectrum of the HVPE samples and downscaled spectrum of a MOVPE template. A high-energy peak above 3.5 eV of unknown origin that shifts to higher energies with higher growth temperatures was observed. Right: Local CL spectra. The high-energy peak originates from the HVPE layer and was not found in the MOVPE layer.



Fig. 8: CL image of the cross-section of the sample grown at 930°C. A clear material contrast is visible between the HVPE layer (top) and the MOVPE layer (bottom).

3. Conclusion

A method to produce semipolar (11 $\overline{2}2$) oriented GaN on r-plane prestructured sapphire substrates was shown.

GaN templates are produced by MOVPE. For our investigations, the MOVPE templates were overgrown by HVPE and compared to the previous MOVPE templates. Finally, a HVPE temperature series was performed to improve the growth conditions.

Compared to the MOVPE template, the FWHMs of the HRXRD rocking curves of the $(11\bar{2}2)$ reflections improved, whereas the $(11\bar{2}4)$ peak got worse. A considerable change in the surface morphology was observed: the sawtooth structure that is common for MOVPE templates turned into a bubble structure with highly reduced roughness.

For further improvements, the growth temperature of the HVPE layer was increased. Only minor improvements of the low growth rate were achieved at the expense of crystal quality. However, it could be shown that the surface roughness decreases with higher temperatures.

Two further phenomena, which are unexplained yet, were observed: on all HVPE samples, huge hillocks emerged. For later device processing, their formation must be avoided. Also a high-energy peak of unknown origin above 3.5 eV was observed in PL and CL measurements. Local CL spectra revealed that this peak belongs to the HVPE layer. Its origin needs further investigations.

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