Establishing a Two Step FACELO Process in HVPE

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In order to reduce the effort needed to create self-separated, freestanding gallium nitride (GaN) layers by thick growth in hydride vapor phase epitaxy (HVPE), we established a two-step facet-controlled lateral overgrowth (FACELO) process in HVPE. Just as for the metalorganic vapor phase epitaxy (MOVPE) FACELO process, the template is produced by MOVPE growth directly on sapphire. This initial GaN layer is masked with 200 nm of silicon nitride (SiN_x), hexagonally structured by optical lithography. Instead of a previously used second MOVPE growth step, the masked sample is directly overgrown in HVPE to create a network of inverse pyramidal structures, before being planarized in a second growth step within the same growth run. Separation of a full 2 inch wafer could be achieved by adding a third, long growth step in HVPE which produced the necessary GaN layer thickness.

1. Introduction

Becoming an increasingly common material, gallium nitride (GaN) still suffers from the lack of suitable substrates. This leads to the need for heteroepitaxy in mid and low price applications. Although homoepitaxy is used for producing blue and green laser diodes, available GaN substrates are much too costly for growing LEDs and transistors on them. To achieve affordable homoepitaxial substrates, great efforts have been made in the field of ammonothermal growth [1]. Yet, thick layers grown by HVPE are still a good candidate to provide GaN wafers for future industrial processes [2, 3]. Although being researched for some time, producing such layers is still no trivial task. Differences in the thermal expansion coefficients of GaN and sapphire and the large mismatch in their respective lattice constants result in strain and defects in the GaN layer and in big curvatures of the respective samples. In order to cope with these problems, several methods have been developed to remove the GaN layer from the substrate such as laser-lift-off (LLO) [4], mechanical polishing [5] or growth on etchable substrates like ZnO [6]. Our approach is to use the difference in thermal expansion coefficients to induce a self separation of layer and substrate during cooldown. With our self-fabricated MOVPE templates, we create weak interlayers which define a breaking point for the separation at a predetermined position. In the last years, our institute has developed a standardized procedure to create suitable templates [7].

However, with two MOVPE growth steps and one subsequent long HVPE growth step for self separation, this procedure is complicated and tedious. To simplify the process, we intend to move the second MOVPE growth step, a FACELO overgrowth of the separation mask, to the HVPE process. The FACELO process contains at least two growth stages,

which needed to be established in the HVPE. The first stage consists of a 3D growth of pyramidal structures, starting from the masked template. This is necessary to bend the defects that protrude from the template through the mask openings. In consequence these defects are expected to run parallel to the wafer surface when the second step of the FACELO growth is applied. In this second step the sample is planarized by growing in a 2D growth mode. Using such a FACELO process, it is possible to reduce the amount of defects in the following epitaxial layers considerably [8]. FACELO has been established for HVPE before [9], however in our case the mask additionally serves as the above mentioned predetermined breaking point for self separation of the thick HVPE layer.

2. Growth Start and 3D Growth Step

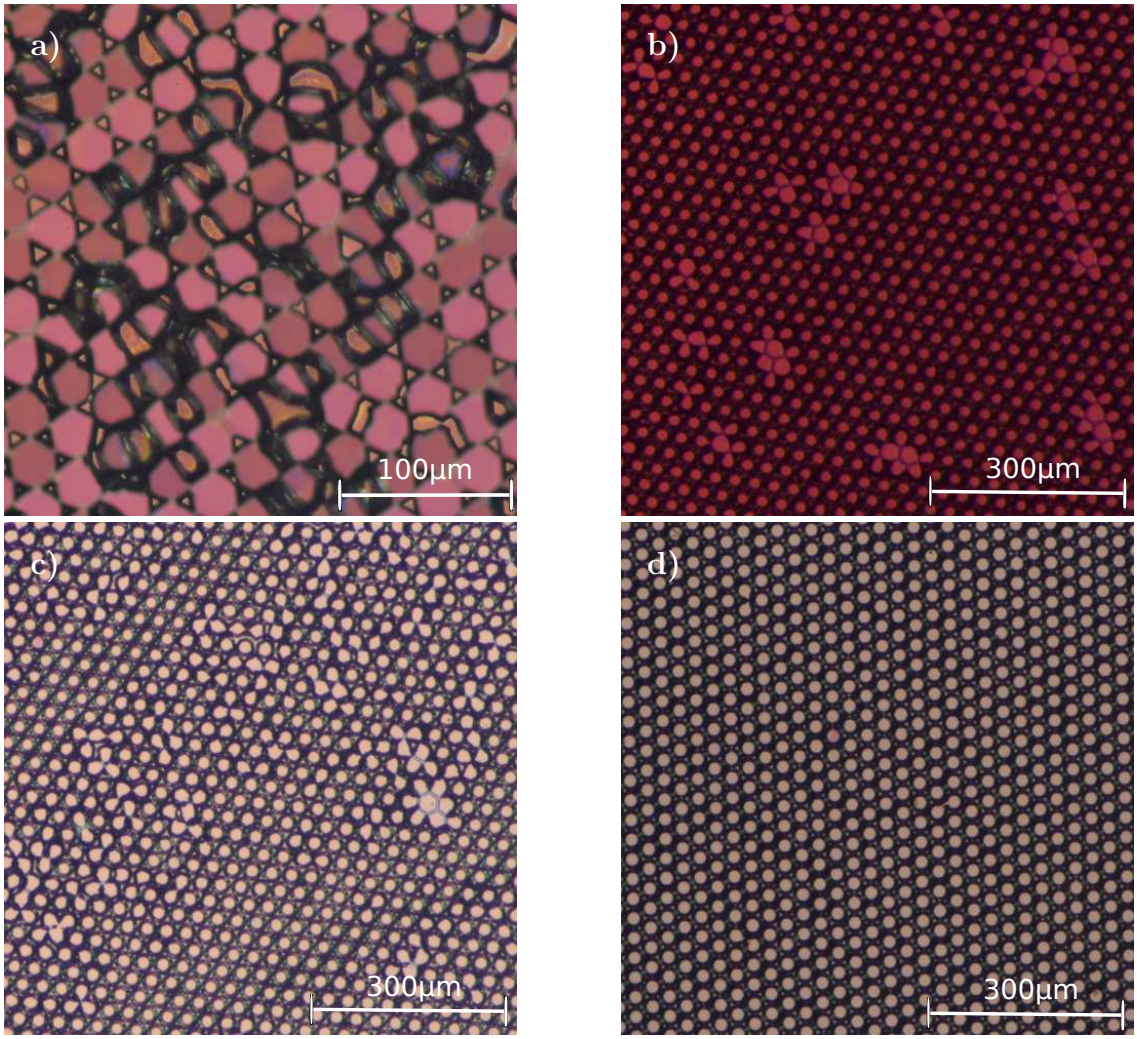

Fig. 1: Nomarski microscopic image, 10 minutes growth on masked template at a) 1050 °C, b) 1000 °C, c) 980 °C and d) 950 °C.

Following the guidelines from a work of Chelda-Gourmala et al. [10], we found suitable

growth conditions for the 3D-mode growth. When comparing the results of this work to our growth result, we see that the V-III ratios can't be taken one to one. Apparently in our reactor a large amount of ammonia doesn't reach the growth zone so that we have to increase the V-III ratio roughly by a factor of 10 for our growth. This brought us to the conclusion that the V-III ratio of 77 which we typically use for thick layer growth (see table 1) would be a good starting point. In order to achieve more homogenous growth at low growth rates and to overcome the design problems of a horizontal reactor with type III showerheads, we increased the V-III ratio to 100. As the first experiment delivered very nicely developed shapes in some spots all over the wafer, we concluded that the V-III ratio was chosen correctly. However, Fig. 1 a) shows a microscopic image of a part of this sample where the growth result was not yet satisfactory. As we can see, growth is limited to some small areas, mainly in the junctions of the mask openings. This indicates high mobility of the source atoms on the wafer, resulting in too strong selectivity.

To decrease the mobility we decreased the growth temperature of the first growth step in three consecutive samples from 1050 °C to 950 °C. Figure 1 b) shows a microscopic image of a typical part of the sample grown at 1000 °C. Growth is now a lot more uniform than at a growth temperature of 1050 °C. However, we still see areas where no growth occured, typically around a small flaw in the mask or template, visible at higher magnifications.

In the next step we decreased the growth temperature to 980 °C (Fig. 1 c)). Now, the amount of spots with no growth has been vastly reduced. Even so, some areas still show distorted features.

Reducing the temperature further down to 950 °C, removes these distortions. Growth is now very homogenous on the whole wafer. A typical microscopic image of such sample can be seen in Fig. 1 d).

3. 2D Growth Step and Planarization

In order to planarize the GaN-layer, we need to enhance the growth parallel to the substrate surface. According to Chelda-Gourmala et al. [10] decreasing the V-III ratio further could highly favor lateral over vertical growth on the $\{1\bar{1}00\}$ facets while favoring vertical over lateral growth on $\{11\bar{2}0\}$ facets. If we increase the V-III ratio, this publication predicts that we get no growth at all on $\{11\bar{2}0\}$ facets while equalizing growth speeds of lateral and vertical growth on $\{1\bar{1}00\}$ facets. As we have a mixture of both facets we couldn't be sure which growth will dominate. Therefore we started two experiments, one growth run reducing the V-III ratio to 50 (Fig. 2 a)), one increasing it to 230 (Fig. 2 b)). As can easily be seen, with the V-III ratio of 50 the inverted pyramids show almost no coalescence. With a V-III ratio of 230, however, we achieve a closed layer with some remaining holes.

To push the growth even stronger into 2D-mode, we increased the V-III ratio to 300 (Fig. 2 c)). Now, at least in the center of the wafer almost all holes are closed after 60 minutes of growth.

The next challenge was to find the right lenght of time to have the top-layer coalesced on the whole wafer with minimal top-layer thickness. From earlier experiments we know that

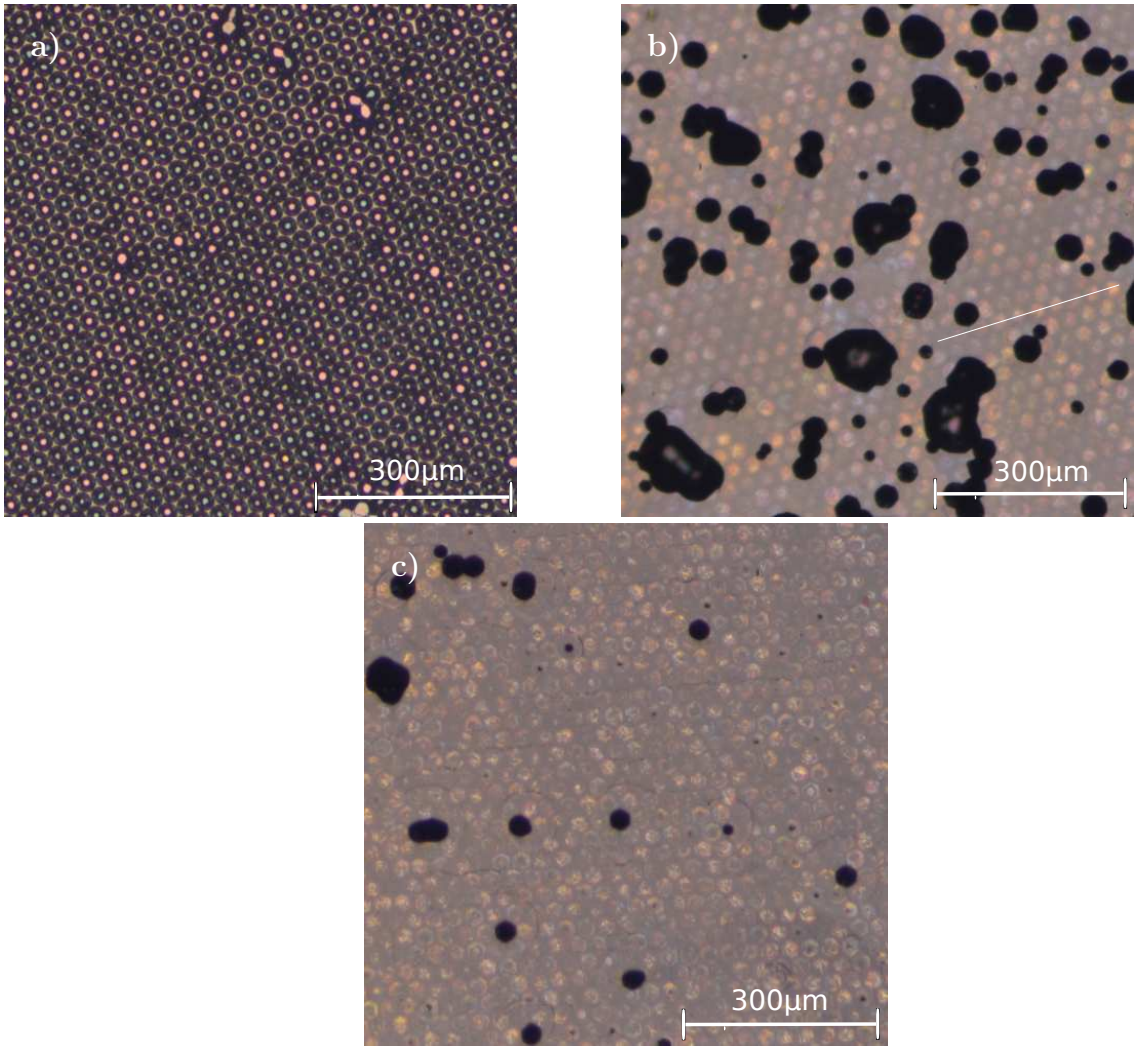

Fig. 2: Nomarski microscopic image, 60 minutes growth on inverted pyramids, wafer center, V-III ratio of a) 50, b) 230 and c) 300.

thick layers grown with this V-III ratio tend to crack. On the other hand the growth-speed is lower at the wafer edge, so this growth has to be applied long enough to coalesce the layer even at the wafer edges. In our experiments, coalescence of the hexagonally shaped mask with 30 µm period and 2 µm openings can be achieved after about 70 µm of vertical growth.

In Figs. 3 a) and 3 b) we can see microscopic images taken from the edge of the wafer after 60 minutes and 90 minutes of growth with a V-III ratio of 230. To be sure that the sample is properly planarized before thick layer growth, we extended the growth in 2D mode to 105 minutes at a V-III ratio of 300.

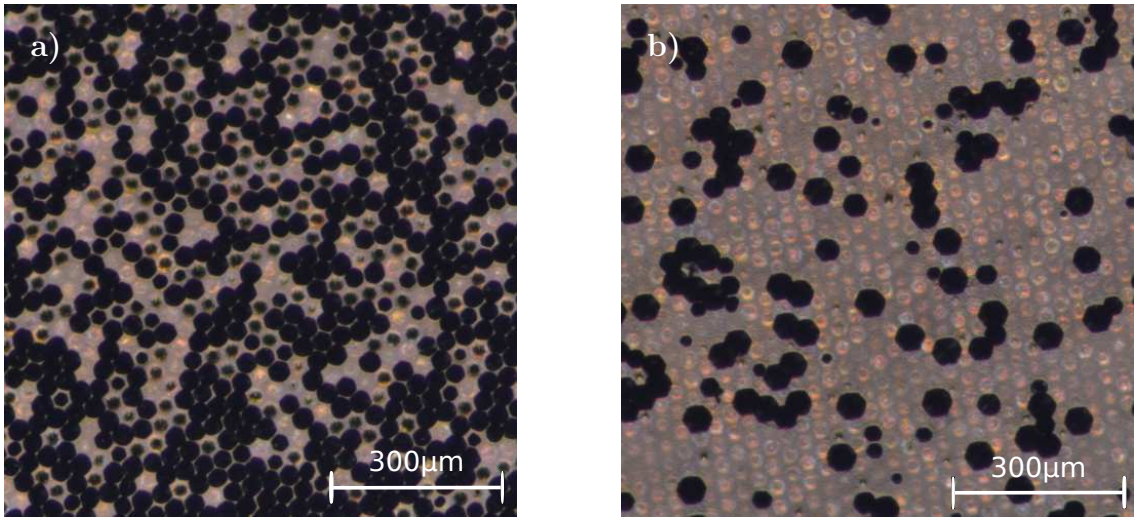

Fig. 3: Nomarski microscopic image, V-III ratio of 230 on inverted pyramids, wafer edge, growthtime a) 60 and b) 90 minutes.

4. Thick Growth and Self-Separation

Applying our standard growth parameters (see table 1), we increased the total layer thickness to enable self-separation during the cool-down-phase of the process. The maximum achievable thickness in our reactor is about 1.5 mm before the sample surface deteriorates. The minimum thickness for samples to separate is about 300 µm. In order to guarantee separation and controle material wear in the reactor we targeted a total thickness of about 700 µm. With a top-layer growth time of 5 h, we achieved a total thickness of 670 µm.

Fig. 4: Photography of whole self-separated, freestanding GaN-wafer.

The thick GaN layer separated in one piece as can be seen in Fig. 4. During growth, some cracks appeared in the material, but could be overgrown in the following process. The

sapphire substrate broke into pieces after separation of the top layer.

Table 1: Growth parameters.

Growth step	Temp. (°C)	NH_3 (sccm)	HCl over Ga (sccm)	Time (min)
3D Growth	950	1000	10	10
2D Growth	1050	3000	10	105
Standard Growth	1050	2300	30	300

In Fig. 5, we see a curvature profile, measured by x-ray diffraction on a line parallel to the wafer flat, through the wafer center. Here we can see the effect of the curvature controle in the template as described in [11]. The minimum curvature measured is 230 km^{-1}, which is well below the value for non curvature controlled samples with a thickness of 670 µm, which lies at about 650 km^{-1}.

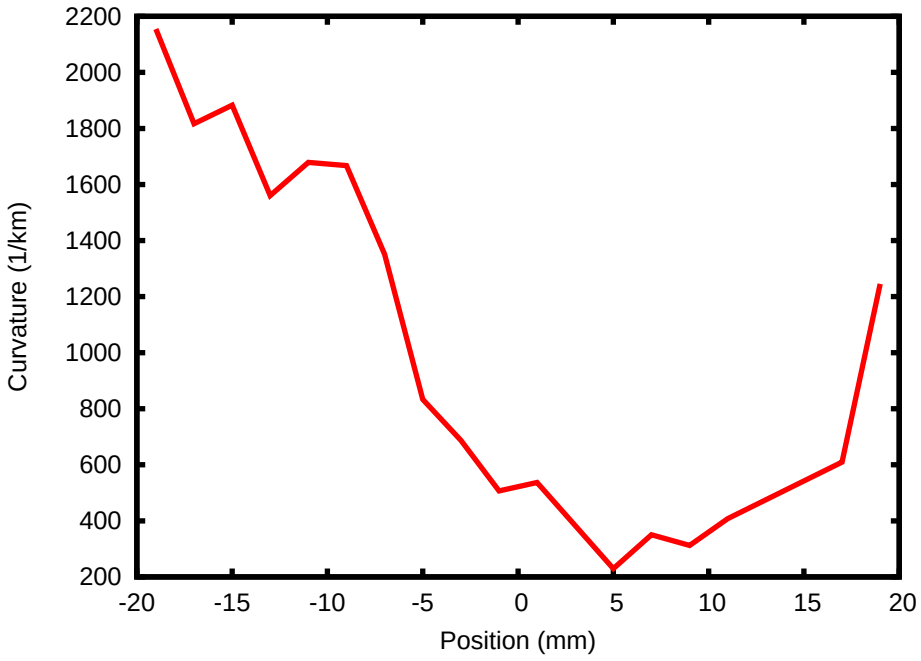

Fig. 5: Curvature profile of whole self-separated, freestanding GaN-wafer.

Low temperature photoluminescence (PL) of a samples grown with this technique shows a full width at half maximum (FWHM) of the D^0X emission of 1.6 meV (Fig. 6). X-ray diffraction (XRD) rocking curve FWHM of the 002 GaN peak can be fitted to 16 arcseconds at the point of the lowest curvature for the full 2 inch freestanding sample shown in Fig. 5. The XRD measurement has been carried out with an open detector and an incoming beam size of 1 mm by 0.05 mm.

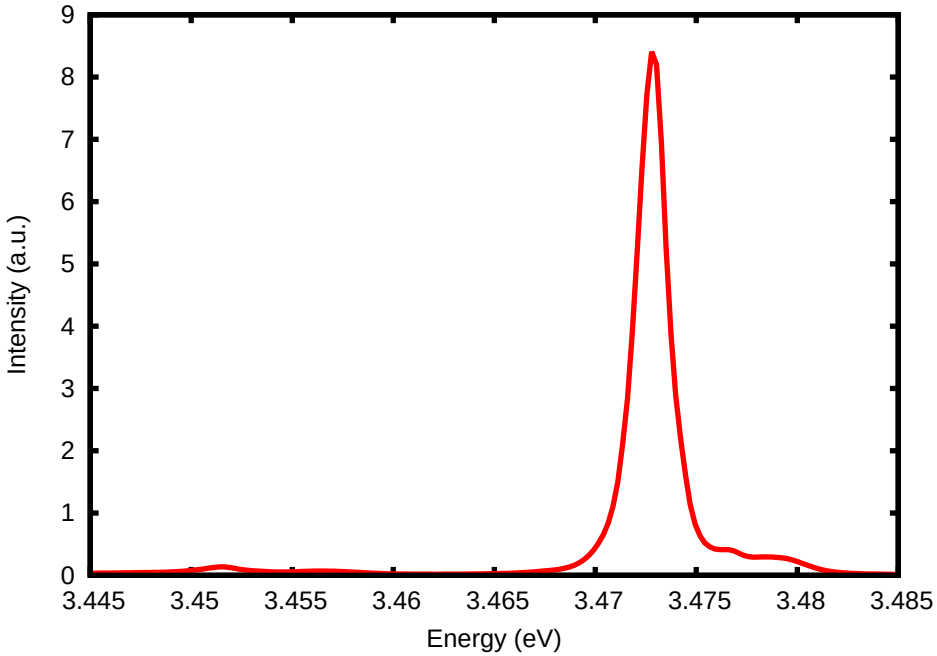

Fig. 6: Low temperature photoluminescence at 14 K of self-separated, freestanding GaN-layer.

5. Summary

We were able to find working growth conditions to overgrow a silicon nitride mask with 3D structures in the HVPE process. Next, we established a process to planarize these three-dimensional structures to regain a flat surface with a low defect density. By increasing the thickness of this template, we were able to get a self-separated, freestanding GaN-layer. With this process we are able to reduce the complexity of the self-separation process as only one MOVPE step is remaining to create the template. Additionally, stable self-separation could be achieved, which has not been the case with the intermediate MOVPE growth-step.

Acknowledgment

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