Structured SiN-masks for Self Separation of Full 2"-GaN Wafers by Hydride Vapor Phase Epitaxy

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Using a dielectric mask structured by optical lithography, freestanding 2"-GaN wafers were prepared by hydride vapor phase epitaxy (HVPE) and self separation during cooldown. The mask was deposited on a GaN template grown on sapphire by metal organic vapor phase epitaxy (MOVPE). We found that the instability of the SiN mask at growth temperature supports the further self-separation. Testing different mask geometries, a hexagonally shaped pattern with a period of 30 µm and an opening of 3 µm showed best performance. This mask allowed the growth and separation of a full 2" GaN wafer by utilizing the stress arising during cooldown from thermal mismatch to the substrate. The thickness inhomogeneity is below 10 % and the samples show good surface morphology.

1. Introduction

Unlike other established semiconductor material systems, nowadays GaN technology is based on heteroepitaxy on foreign substrates due to missing GaN bulk crystals. While the ammonothermal growth showed good progress for the fabrication of these demanded substrates in the last years [1,2], hydride vapor phase epitaxy (HVPE) is still considered as the most promising tool. Nevertheless HVPE growth of thick GaN samples is a challenge. It is mostly a heteroepitaxial process, where large strain is generated on one side by lattice mismatch of the foreign substrate material to GaN and on the other side by the large mismatch of the respective thermal expansion coefficients yielding to severe bowing and often to strong cracking of the grown samples. In order to reduce strain and bowing in the final quasi-substrate, a removal technique of the foreign substrate from the grown GaN layer is required. Many approaches towards that challenge were reported in literature, such as laser-lift-off (LLO) [3], mechanical polishing for substrate removal [4] or growth on etchable substrates like GaAs or ZnO [5, 6]. A self-evident technique is the use of stress arising during cooldown for the separation. Many publications describe such an approach [7, 8], nevertheless it is a difficult process as it requires some kind of interlayer in the epitaxial structure for separation. On one hand, such a layer should weaken the interface, on the other hand, most of the stress during cooldown should arise exactly at this position, otherwise the cracking may occur at an undefined position in the crystal. And finally there is the need to find a compromise of weak connection and good material quality in the GaN grown above the interlayer. This seems to be very critical e.g. for low temperature interlayers [9].

In this work we used dielectric masks, structured by optical lithography and dry etching by an ex-situ process. This idea was originally used for defect reduction by epitaxial

lateral overgrowth (ELOG) being nowadays fairly well established, and is also applied for self-separation [10]. This method is well reproducible, in particular compared to most of the in-situ deposited interlayers. The material quality of the overgrown layer above the mask is excellent and the method also allows full control on the mask pattern and hence on the strength of the connection.

2. Experimental

The HVPE growth was performed in a commercial Aixtron single-wafer HVPE system with a horizontal quartz-tube, heated in a furnace with five zones. A 1:1 mixture of nitrogen and hydrogen was used as carrier gas, as it showed best performance regarding cracking [11]. Ammonia was applied as nitrogen precursor, while for the group-III element GaCl was used, formed inside the reactor by streaming HCl gas over a liquid Ga source heated to 850 °C. The used showerheads for the GaCl injection together with the carrier gas flows were carefully adjusted to reduce parasitic deposition as much as possible. The temperature and pressure for the GaN growth were kept constant at 1050° C and 900 hPa respectively. In order to improve the surface morphology, the V/III-ratio, pressure and growth rate were changed during the last few micrometers of the growth. Details of this procedure can be found elsewhere [12]. The growth was started at a fairly low growth rate of about 10 µm/h with a high V/III-ratio to pronounce lateral growth and then changed up to 100 µm/h.

The HVPE-growth was performed on 2 μm thick GaN template layers grown in an Aixtron 200/4 RF-S MOVPE system on (0001) sapphire wafers with a thickness of 430 µm and a diameter of 2". The substrates had a miscut of 0.3° towards the a-plane, yielding to an improved surface morphology [12]. For defect-reduction an in-situ SiN-layer was deposited during the growth of the GaN template [13]. Such templates are strongly compressively strained and allow crack-free growth of comparably thick HVPE layers more easily. On these templates, a 200 nm thick SiN or SiO_2 layer was deposited by plasma enhanced chemical vapor deposition (PECVD) and structured by optical lithography and dry etching. We investigated several different patterns for these studies.

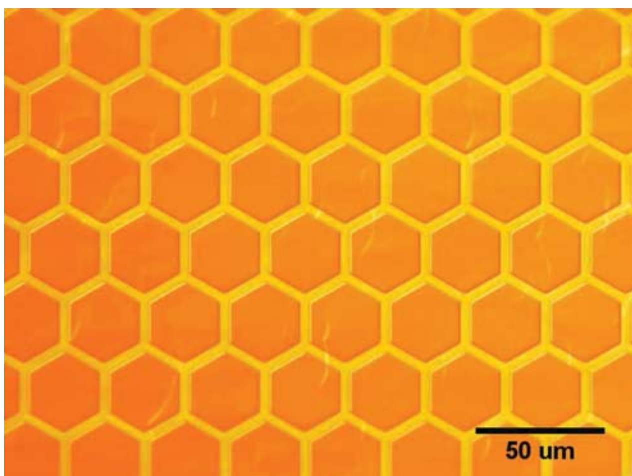

Fig. 1: Honeycomb pattern where the masked area is hexagonally shaped. The period of the mask is varied between 15 µm and 100 µm.

The first one is a simple stripe pattern with masked stripes of a width of 8 μm and an opening of 3 μm in between. The stripes are aligned along the $\langle 1\bar{1}00 \rangle$-direction. The second pattern is hexagonally shaped like a honeycomb with open trenches between masked hexagons (Fig. 1). For this pattern the size of mask and trench were varied, see table 1. Again, the trenches run along the $\langle 1\bar{1}00 \rangle$-direction of GaN.

After locally removing the mask by dry etching, the samples were put back into the MOVPE system to initiate lateral growth starting from the open trenches in the mask partly covering the masked area. This growth step starts at fairly low temperature and low V/III-ratio corresponding to a 3D growth mode. The patterns formed in this step having triangular cross-sections were afterwards overgrown with increased temperature and increased V/III-ratio for a more lateral growth over the masked areas. Although the masked area is not completely closed during this step, it helps to achieve a closed layer at an early stage in the HVPE growth.

3. Results and Discussion

3.1 Mask material

First, the applicability of SiO_2 and SiN as masking interlayer for a subsequent self-separation were investigated. Therefore, two samples were prepared with the same stripe pattern and then overgrown using the same conditions in MOVPE and HVPE with an about 300 μm thick GaN layer. On both samples, self-separation during cool-down could be observed. However, only the one with the SiN-mask separated at the prepared interlayer. Obviously, the SiN mask dissolves during HVPE growth, additionally leading to a dissolution of the nearby GaN, resulting in big cavities in the GaN buffer. In Fig. 2 a SEM-picture of the cross-section of this sample is shown. The position of the former mask can be identified by the big void due to a late coalescence of the overgrown layer. Mainly the GaN buffer below the mask is partly dissolved while the buffer below the former opening is not attacked. In contrast to the sample with SiO_2-mask, horizontal cracks were only observed at the mask position. SiO_2 is much more stable compared to SiN. Hence mask and GaN buffer of the SiO_2 masked sample survive the HVPE growth unharmed. Instead of a separation on the mask, horizontal cracks about 20 μm above developed as consequence of the high stress due to the thermal mismatch of sapphire and GaN during cooldown.

Obviously, the dissolution of the mask together with the GaN nearby is essential for the separation. A sample, where the SiO_2-mask material was removed after the second MOVPE step resulting in cavities where the grown triangular overlaps the former mask due to a less pronounced lateral growth in HVPE, showed the same behavior as for remaining SiO_2. Again, horizontal cracks appeared above the cavities (see Fig. 3). These horizontal cracks also lead to self separation and freestanding GaN, but in a fairly uncontrolled random process. These samples also show several vertical cracks yielding to only small pieces of GaN instead of a full free-standing wafer.

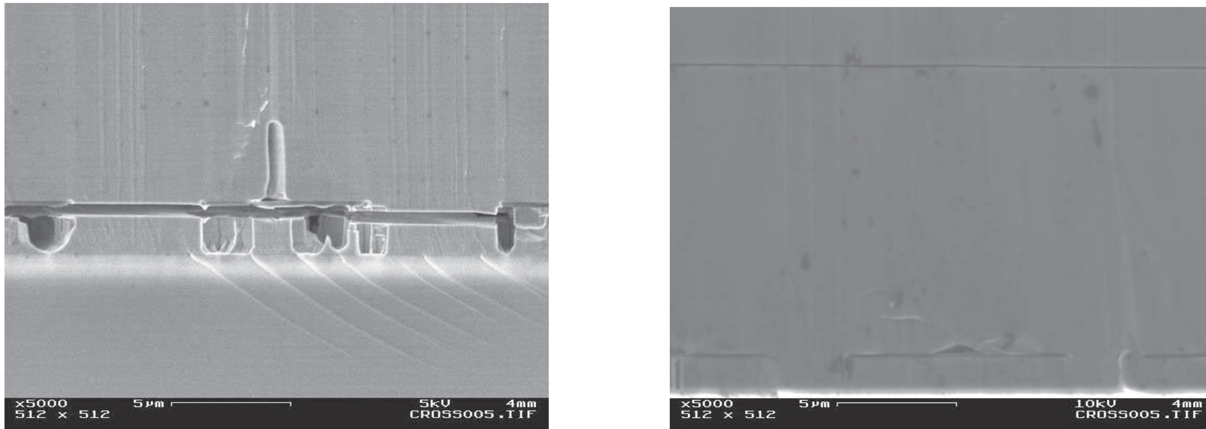

Fig. 2: Cross section SEM pictures after HVPE growth of about 300 µm. The left picture shows the SiN mask which dissolved and also attacked the GaN directly below the mask. The mask position is indicated by the vertical cavity due to a late coalescence of the overgrown GaN. The right picture shows the growth on a SiO_2 mask. The mask material and the GaN nearby is still stable, but a horizontal crack about 20 µm above the mask developed.

3.2 Mask pattern

Freestanding GaN-pieces that were produced with the stripe pattern showed a strong asymmetric (concave) bow with a curvature of $1100\,\mathrm{km}^{-1}$ and $1550\,\mathrm{km}^{-1}$ parallel and orthogonal to the stripes, respectively. In order to suppress this asymmetric bowing, we investigated the honey-comb like pattern as described above (see Fig. 1). In order to optimize the separation properties, the ratio between masked and open area was varied keeping the opening width constant at 3 µm thus changing the filling factor FF, i.e. the ratio of the masked to open area (table 1). We expected a better separation for a larger FF. For a 200 µm thick GaN layer no separation was achieved for the 15 µm period sample. The 30 µm period showed full separation, although the GaN layer broke into several pieces due to the comparably low layer thickness. However, for the larger periods of 60 µm and 100 µm, it was not possible to get a closed layer during the HVPE growth. To increase the FF at still acceptable period length, we have reduced the width of the openings to

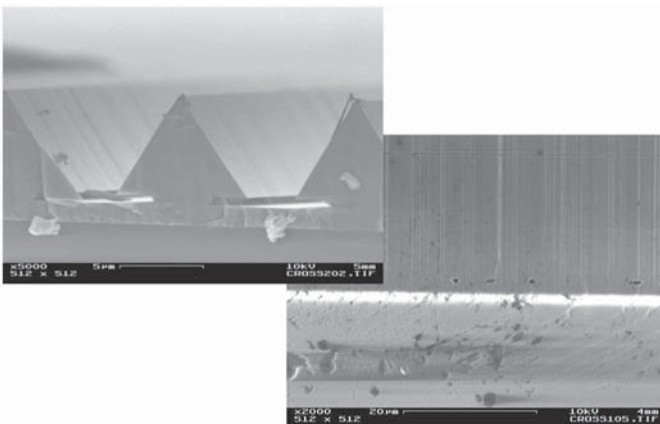

Fig. 3: SEM picture of a sample where the SiO_2 was removed after the second MOVPE step (left). The part, that is overgrown over the former mask will remain as cavity after the HVPE growth. The right picture shows a crosssection after HVPE growth. Similar to samples where the SiO_2 was not removed, horizontal cracks above the interlayer developed and are visible in the SEM picture.

1.5 μm. This lead to a slightly improved separation. However, as this is at the limit of our optical lithography, further experiments were done with 3 μm openings.

We also observed that the separation does not depend on the layer thickness if a thickness of about 200 μm is exceeded. However, a larger thickness is needed to avoid vertical cracking of the GaN layer during separation.

Table 1: Ratio of masked to open area (FF) for different patterns.

Pattern	Trench width	Period	Filling factor (FF)
stripe	3 μm	8 μm	72%
hexagon	3 μm	15 μm	64%
hexagon	3 μm	30 μm	81%
hexagon	3 μm	60 μm	90%
hexagon	3 μm	100 μm	94%
hexagon	1.5 μm	30 μm	90%

3.3 Properties of separated GaN-wafers

In Fig. 4 a photograph of an about 1.5mm thick separated GaN wafer is shown with a thickness inhomogeneity of only 10 %. Such a thickness is actually the limit of our HVPE system due to parasitic depositions inside the reactor. It has a very smooth surface (Fig. 5).

All separated samples exhibit a concave bow depending on the total layer thickness. The bow of the 1.5 mm thick sample was determined by HRXRD measurement to be 277 km^{-1} while a thinner sample with half the thickness showed a bow of 750 km^{-1}.

Fig. 4: Photograph of a full 2” GaN wafer self-separated during cool-down by growth on the hexagonally shaped mask with a period of 30 μm. The thickness of the wafer is about 1.5 mm.

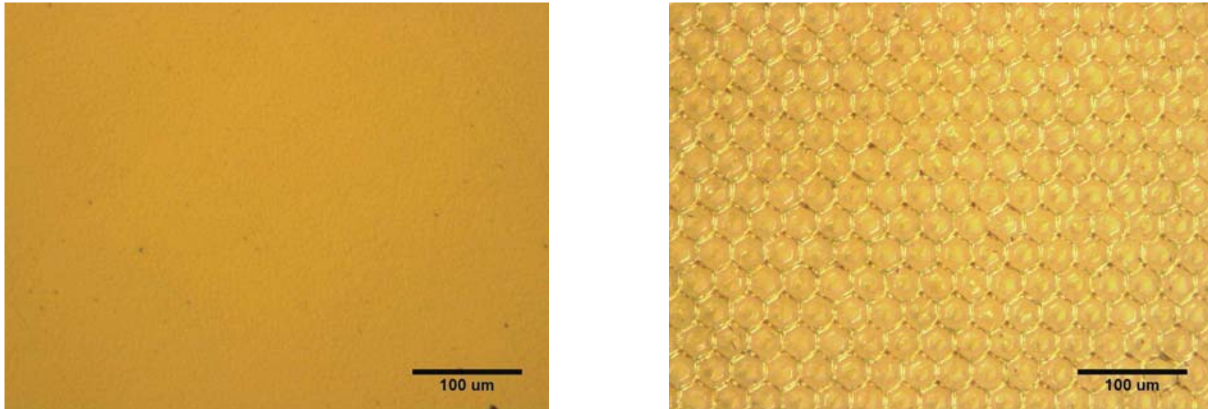

Fig. 5: Optical microscopy photograph with Nomarski contrast of the free-standing self-separated GaN layer. The surface is very smooth (left). On the backside (right), the mask structure is still observable, proving that separation happened on that interlayer.

The quality of the layer was further investigated by PL and HRXRD measurements. The full width at half maximum (FWHM) of the (002)- and (102)-reflections are 75 arcsec and 230 arcsec, respectively.

The position of the donor bound exciton (D^0X) at 3.470 eV measured by low temperature photoluminescence with a HeCd-laser as excitation (Fig. 6) indicates a strain free surface. The FWHM of less than 1 meV for the D^0X shows the good quality. The dislocation density determined by a chemical etching method, where we used hot HCl-gas and atomic force microscopy [14] was below $1 \cdot 10^6\,\mathrm{cm}^{-2}$. By room temperature Hall measurement we determined a carrier mobility of $740\,\mathrm{cm}^2/\mathrm{Vs}$ and a carrier concentration of $6 \cdot 10^{16}\mathrm{cm}^{-3}$.

4. Conclusion

Full 2”-wafers of freestanding GaN of high quality were fabricated by a self-separation process during cool-down by an inserted dielectric mask. It was found that SiN as mask material dissolves at the chosen temperature in HVPE and leads to cavities in the GaN-buffer below, strongly enhancing the self-separation process and making the production of crack-free freestanding GaN wafers possible.

Acknowledgments

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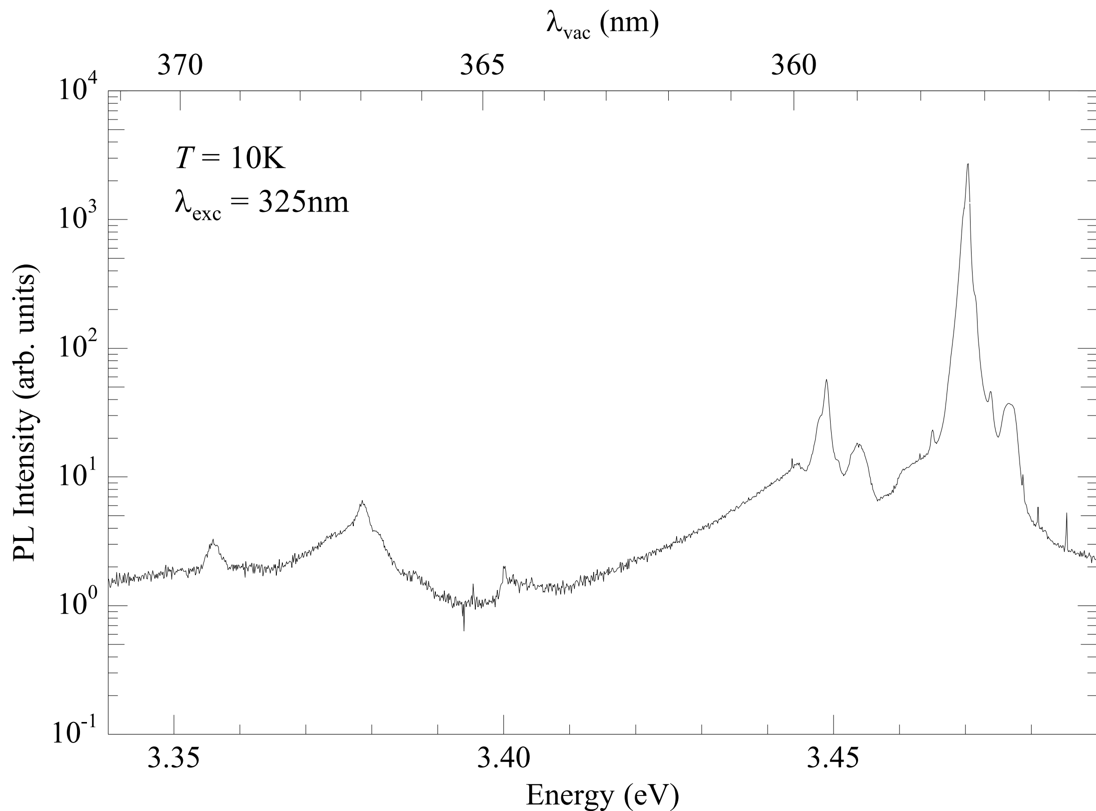

Fig. 6: Low temperature (10 K) PL spectrum of a freestanding sample, grown on the hexagonal mask with a period of 30 μm.

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