Optimization of GaN Based Light Emitters With Semipolar Quantum Wells and Sub-μm Patterning Within the Active Zone

Robert A. R. Leute

The selective growth of sub-micrometer sized GaN stripes running along a-direction on c-oriented GaN templates results in semipolar quantum wells (QWs) grown on the $\{10\bar{1}1\}$ *side facets that can be embedded easily. Therefore we achieve light emitting devices with semipolar quantum wells on large areas with flat surfaces for easy processing. The influence of the growth mask is thoroughly investigated and structural analysis of the embedded QWs is presented.*

1. Introduction

There is a high scientific interest in semipolar GaN crystal planes for efficient long wavelength light emitters [1, 2]. The use of free-standing GaN templates for homoepitaxy is favoured by many groups [3–5]. However, the high cost and small size of commercially available semipolar GaN substrates motivates the pursuit of heteroepitaxial alternatives [6]. Three-dimensional GaN structures, based on growth in c-direction, with semipolar side facets are especially attractive, since they can be grown on large and inexpensive foreign substrates with high crystal quality [7]. For device processing, however, planar surfaces are favored, allowing conventional structuring methods for contacts, resonator formation, etc. This can be achieved by reducing the size of the 3D GaN to a few hundred nanometers and embedding the active, semipolar QWs within c-plane layers. Over the course of the year 2011 a patterning process based on laser interference lithography was established within the Institute of Optoelectronics [8] and first light emitting diodes (LEDs) with embedded GaN stripes were presented. In the following, we present the optimization of the processing. The influence of the growth mask is investigated and structural analysis of the embedded quantum wells by transmission electron microscopy (TEM) and scanning transmission electron microscopy (STEM) is shown. This was partly published in [9].

2. Experimental

All epitaxial growth is done in an Aixtron-200/4RF-S HT MOVPE reactor with standard precursors. Silane and Cp_2Mg are used for doping; Pd diffused hydrogen and high purity nitrogen are used as carrier gases. About 3 μm thick c-oriented GaN templates are grown on c-plane sapphire with conventional growth conditions. We use an oxygen doped AlN nucleation layer as well as an in-situ SiN interlayer to reduce the defect density [10]. The topmost layer of 1.8 µm thickness is silicon-doped to achieve a nominal doping level of $3.5 \times 10^{18}\,\mathrm{cm}^{-3}$.

For laser interference lithography, the beam of a HeCd laser ($\lambda = 325$ nm) is expanded by a lens and spatially filtered with a pinhole. Half of the beam hits the sample directly, the other half is reflected by a mirror, positioned very close to the sample with an angle of about 90°. Both beam halves interfere with each other and create a stripe pattern with a periodicity as low as 230 nm on an area of a semicircle with a diameter of 2.5 cm to 3 cm. The geometry is based on Lloyd's mirror [11]. The alignment of sample and mirror was chosen to result in resist stripes running parallel to the a-axis in GaN. A thin layer (15 nm) of titanium is subsequently deposited onto the structured templates by electron-beam evaporation and the pattern is transferred via lift-off. The templates are then cleaned with an aqueous KOH solution and re-loaded into the MOVPE reactor where the Ti mask is nitridized *in-situ*. Afterwards, the second growth step is carried out, consisting of n-doped stripes with $\{10\bar{1}1\}$ side facets, two InGaN quantum wells, an undoped spacer and finally a p-doped planarization layer. The epitaxial structure includes neither an electron blocking layer nor an InGaN pre-well common to c-plane LEDs. The stripes are grown for 120 s at 950°C with a V/III ratio of 260. The QWs are grown at 760°C with TEGa and nitrogen as carrier gas for 460 nm emission wavelength. The spacer is grown first at 950°C, then temperature is increased to 1000°C. For the embedding p-GaN layer the temperature was varied from 1000°C to 1090°C while the V/III ratio was kept at 1080.

For electrical characterization, the LEDs were annealed in an ambient atmosphere at 750°C for 60 s to activate the Mg-acceptors, then 1 µm thick circular indium contacts were evaporated onto the p-side of the LED.

2.1 Optical characerization and influence of mask material

The devices exhibit a highly polarized light emission (shown in Fig. 1) due to the semipolar character of the quantum wells and the regular arrangement of the stripes [12], however the intensity is rather low.

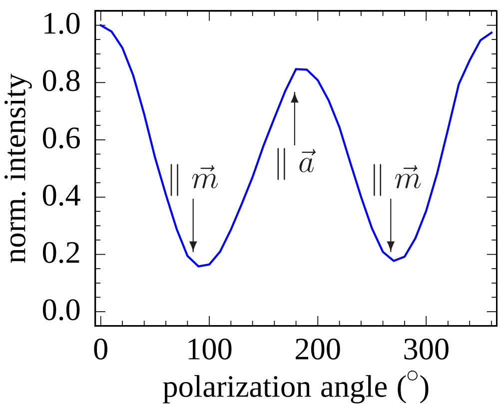

Fig. 1: Polarization dependent measurement of the emission from the semipolar QWs. Polarization angle is given relative to the orientation of stripes.

Transmission electron microscopy (TEM) measurements were performed by J. Thalmair at the University of Regensburg in the group of Prof. J. Zweck to investigate the reasons for the low light output. Figure 2 shows a TEM picture of the embedded active zone with well-developed QWs of 3 nm thickness. Figure 3 however reveals that in some areas c-plane surfaces exist with QWs which are over 10 nm thick, which would result in a nominal emission wavelength as long as 800 nm if we assume an identical indium content. Nonradiative recombination dominates in these thick c-plane QWs due to the intrinsic piezoelectric field and hamper the performance of the LED.

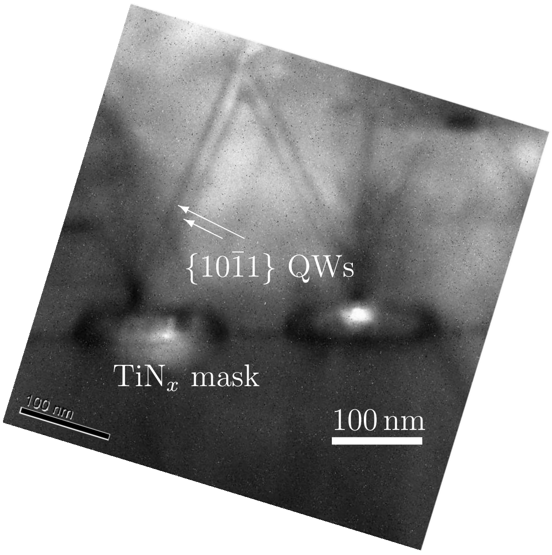

Fig. 2: TEM cross section of the embedded active region with well developed semipolar quantum wells.

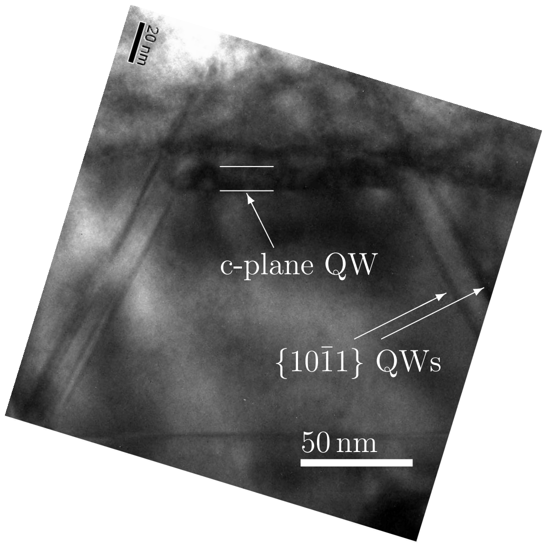

Fig. 3: TEM cross section view of a region where adjacent stripes coalesced and formed a c-plane surface. The c-plane QW is enlarged.

In order to exclude the influence of c-plane QWs, reference samples were structured by electron-beam lithography with a honeycomb pattern. The resulting sub-micrometer sized inverse pyramids have $\{10\bar{1}1\}$ side facets, too, and show almost no c-plane surfaces at all within the patterned area. All growth parameters were kept identical to the stripe samples. A planarized LED with such structures within the active region showed an improved output power, see Fig. 4, corroborating the negative effect of the c-plane areas.

The immediate vicinity of the TiN_x mask to the active zone of the LEDs further influences the device performance. We assume that TiN_x absorbs substantial fractions of the light created within the active region of the LED as well as increases leakage currents due to its metallic character. Ex-situ removal of the TiN_x mask after the growth of the 3D GaN stripes with a mixture of hydrogen peroxide and sulfuric acid resulted in a tenfold increase of the output power and a decrase of the leakage current at reverse bias, compared to a similar sample where the TiN mask remained embedded. Therefore, efforts were undertaken to replace the TiN_x mask by a dielectric SiO_2 or SiN_x mask. Patterning via lift-off of sputtered SiO_2 proved impractical and was replaced by pattern transfer through dry etching with a fluorine containing plasma.

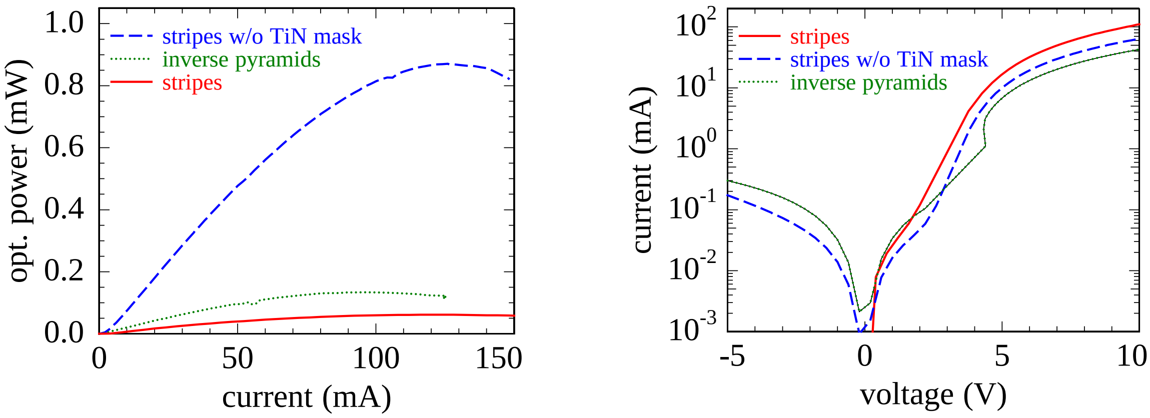

Fig. 4: Power–current (left) and current–voltage (right) characteristics for semipolar LEDs with embedded 3D structures. A substantial increase of power was achieved by embedding inverse pyramids. A huge increase was achieved by ex-situ removal of the TiN_x mask.

2.2 Structural analysis

LEDs with embedded sub-µm stripes based on a dry etched SiN_x growth mask were investigated at the Central Electron Microscope Facility of Ulm University headed by Prof. U. Kaiser. TEM and STEM measurements were performed by J. Biskupek.

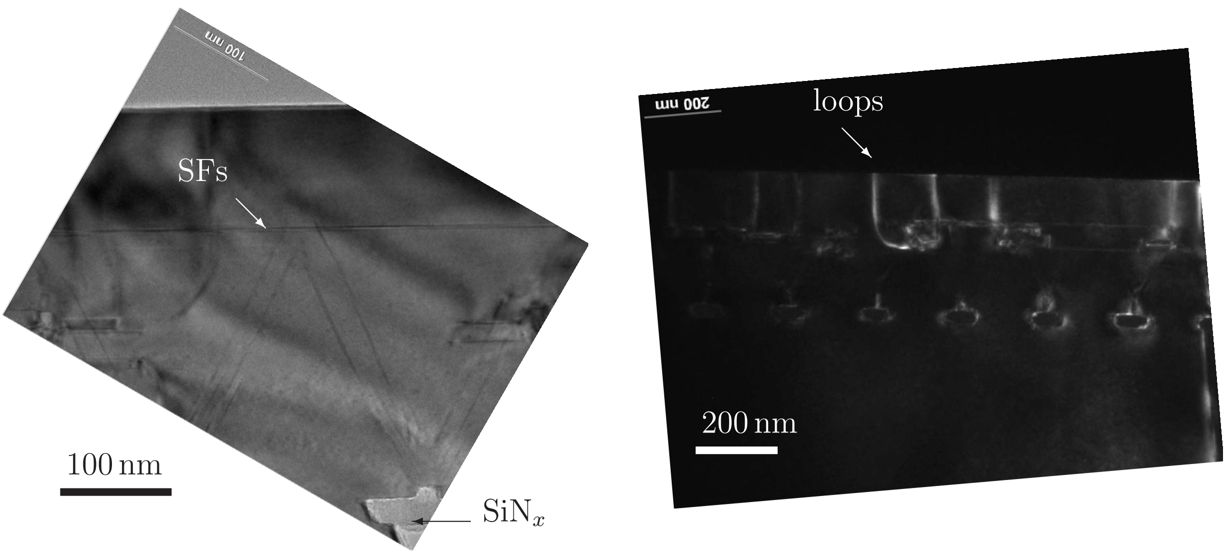

Fig. 5: TEM micrographs of embedded sub-µm sized GaN stripes with semipolar QWs. Stacking faults (SFs) originate at the ridges (left) and can lead to dislocation loops penetrating to the surface (right).

We observe the creation of stacking faults (SFs) at the ridges of the stripes which can cause dislocation loops penetrating to the surface, shown in Fig. 5. We assume that strain accumulates at the ridges during quantum well growth which is relaxed by the formation of stacking faults. The quantum wells themselves show an inhomogeneous thickness distribution and especially the later grown quantum well is not fully formed along the facet, as can be seen in Fig. 6. We tentatively correlate the inhomogeneity of the quantum wells with the low homogeneity of the mask patterning still present when using the laser interference lithography.

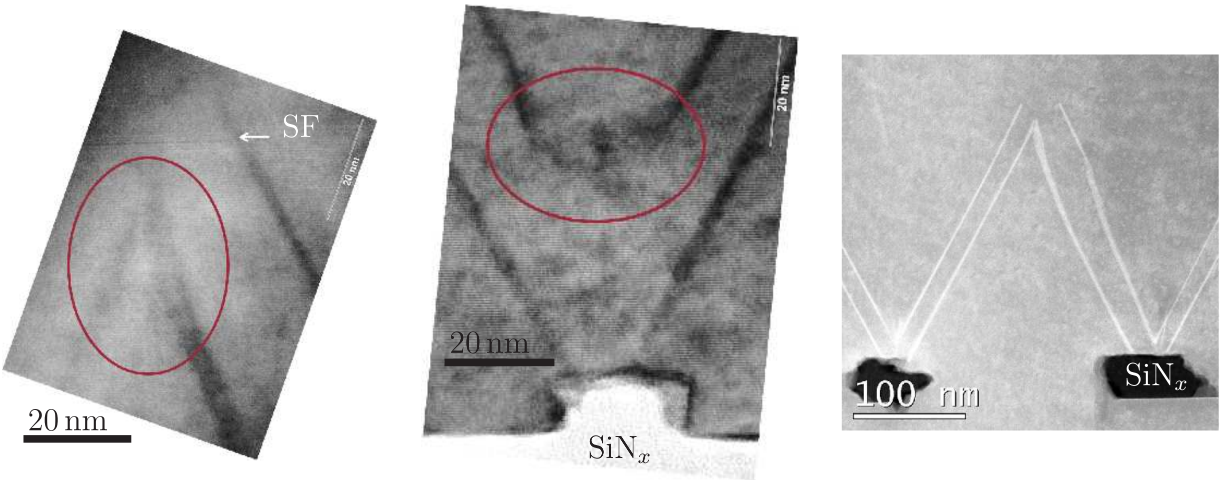

Fig. 6: TEM (left and middle) and STEM (right) micrographs of embedded sub-µm sized GaN stripes with semipolar QWs. The later grown quantum well is not fully formed during growth — especially at the top and bottom of the stripes (left and middle). An inhomogeneous mask patterning might cause severe QW inhomogeneity (right).

3. Conclusion and Outlook

LEDs with semipolar QWs have been manufactured and characterized. A strong influence of the embedded TiN$_x$ mask was found leading to its replacement with SiN$_x$. Structural analysis revealed thickness inhomogeneities of the InGaN QWs and the creation of stacking faults at the ridges of the embedded sub-µm stripes.

In order to improve the homogeneity of the patterning, subsequent work will employ nanoimprint patterning with first experiments already ongoing. In order to prevent the formation of stacking faults, only a single quantum well will be used for future samples and an InGaN prewell is included to mitigate the abruptness of the strain gradient.

Acknowledgment

Technical and scientific support by S. Kizir, I. Schwaiger, J. Wang, R. Blood, D. Heinz, T. Meisch and R. Rösch is gratefully acknowledged.

References

[1] M.T. Hardy, D.F. Feezell, S.P. DenBaars, and S. Nakamura, “Group III-nitride lasers: a materials perspective”, *Materials Today*, vol. 14, pp. 408–415, 2011.

[2] J. Han and M. Kneissl, “Non-polar and semipolar nitride semiconductors”, *Semicond. Sci. Technol.*, vol. 27, pp. 020301-1–2, 2012.

[3] Y. Enya, Y. Yoshizumi, T. Kyono, K. Akita, M. Ueno, M. Adachi, T. Sumitomo, S. Tokuyama, T. Ikegami, K. Katayama, and T. Nakamura, “531 nm green lasing of InGaN based laser diodes on semi-polar $\{20\bar{2}1\}$ free-standing GaN substrates”, *Appl. Phys. Express*, vol. 2, pp. 082101-1–3, 2009.

[4] A. Avramescu, T. Lermer, J. Müller, C. Eichler, G. Bruederl, M. Sabathil, S. Lutgen, and U. Strauss, “True green laser diodes at 524 nm with 50 mW continuous wave output power on *c*-plane GaN”, *Appl. Phys. Express*, vol. 3, pp. 061003-1–3, 2010.

[5] R.M. Farrell, E.C. Young, F. Wu, S.P. DenBaars, and J.S. Speck, “Materials and growth issues for high-performance nonpolar and semipolar light-emitting devices”, *Semicond. Sci. Technol.*, vol. 27, pp. 024001-1–14, 2012.

[6] F. Scholz, “Semipolar GaN grown on foreign substrates: a review”, *Semicond. Sci. Technol.*, vol. 27, pp. 024002-1–15, 2012.

[7] T. Wunderer *et al.*, “Three-dimensional GaN for semipolar light emitters”, *Phys. Status Solidi B*, vol. 248, pp. 549–560, 2011.

[8] R.A.R. Leute and D. Heinz, “Sub-μmpatterning for semipolar GaN based light Emitters”, *Annual Report 2011*, pp. 41–46, Ulm University, Institute of Optoelectronics.

[9] R.A.R. Leute, D. Heinz, J. Wang, F. Lipski, T. Meisch, K. Thonke, J. Thalmair, J. Zweck, and F. Scholz, “GaN based LEDs with semipolar QWs employing embedded sub-micrometer sized selectively grown 3D structures”, *J. Cryst. Growth*, http://dx.doi.org/10.1016/j.bbr.2011.03.031, 2012.

[10] J. Hertkorn, F. Lipski, P. Brückner, T. Wunderer, S.B. Thapa, F. Scholz, A. Chuvilin, U. Kaiser, M. Beer, and J. Zweck, “Process optimization for the effective reduction of threading dislocations in MOVPE grown GaN using in situ deposited SiN_x masks”, *J. Cryst. Growth*, vol. 310, pp. 4867–4870, 2008.

[11] H. Lloyd, in *Proceedings of the Royal Irish Academy 1836-1837*, p. 6, Royal Irish Academy, 1837.

[12] M. Feneberg, F. Lipski, R. Sauer, K. Thonke, P. Brückner, B. Neubert, T. Wunderer, and F. Scholz, “Polarized light emission from semipolar GaInN quantum wells on $\{1\bar{1}01\}$ GaN facets”, *J. Appl. Phys.*, vol. 101, pp. 053530-1–6, 2007.