Advancement of the Core-Shell-Structure Epitaxy Process for InGaN-Nanorod-LEDs on Si (111)

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Introduction

The III-nitride semiconductors and their ternary compounds hold direct band gaps which cover the total visible spectrum. However, III-nitride-LED fabrication faces several obstacles: up to now there is no cost-efficient way of growing high quality III-nitride substrates so that growth of LEDs takes place on foreign substrates. Growth on foreign substrates leads to lattice defects and thus to a decrease of device performance [AMB98, DIK06]. The most preferred substrate, sapphire, is an insulator which renders device contacting difficult. Furthermore, areal metalorganic vapour phase epitaxy growth of III-nitride layers is very expensive due to high amounts of the high priced precursor materials. At last, III-nitrides contain intrinsic electrical fields in c-direction attributed to the different electronegativity of the metal atoms and the nitride atoms leading to the so called quantum confined stark effect, causing band bending in the quantum well which separates the electron- and hole-wave functions, decreasing the recombination efficiency and being responsible for a red-shift in light emission [TAK97]. Due to the anisotropy of III-nitrides, growth can occur in two directions called nitride- and gallium-polar. Mixed polarity causes inversion domain boundaries (IDBs) with a high amount of defects. Nanorods are an appropriate alternative to layer based-LEDs due to their well-known advantages: they provide a high area to volume ratio and thus are far cheaper in production. Charge carrier transport in the non-polar m-direction prevents the quantum confined stark effect [CHE13, CHE14]. Also, the nanorod structure leads to a reduction of lattice tension, enabling the growth of high-quality-GaN on n-conducting silicon (111) substrates, providing a substrate contact without additional processing steps. The Solid-State Electronics Department in Duisburg showed the growth of high-speed nanorod-on-Si-GaN-LEDs [KOE15]. However, these LEDs lack in electroluminescence performance due to yellow luminescence and high growth inhomogeneity, enabling only a small rate of all nanorods to contribute to lighting. Further challenges are the mixed polarity growth, low overall number of grown nanowires and inhomogeneous InGaN-growth. In this work we will discuss the optimization of polarity, density, homogeneity and the core-shell growth of those nanorods.

Experiment

Unipolar growth of GaN is necessary for high quality LEDs. Better conductivity and less yellow luminescence make N-polar growth favourable [DU11, MAC02]. To examine the polarity, kaliumhydroxide- (KOH) etching has been widely promoted, etching N-polar areas and leaving Ga-polar areas unaffected. Two approaches have been made to reduce the number of IDBs: first, the predeposition before AlN-layer-growth has been varied and the AlN layer as well as the GaN-rods have been investigated by KOH-etching and SEM. Predepositions of aluminium (Al) and nitridation in different times have been executed by providing only Al- or nitride-precursors. Even though in this way no unipolarity could be achieved, the number of
IDBs is highly dependent of the predeposition and could be reduced to a minimum by using an Al-predeposition time of 15 s.

![Fig.1](image)

**Fig.1** a) GaN-nanorod after 5 minutes of KOH-etching (2 mol/l, 50° C). Ga-polar regions show a pyramidal shape and resist KOH-etching whereas N-polar regions favour flat c-facets and are dissolved in KOH. No predeposition. b) GaN-nanorod after KOH-etching, 15s Al-predeposition.

The second approach for unipolar nanorods is to utilize the high difference in growth speed leading to an overgrowth of one polarization. Hydrogen (H) –radicals cause the formation of Ga-H complexes and thus the etching of Ga on Ga-terminated surfaces. In Ga-polar nanorods, the c-facet is Ga-terminated [LI11], leading to an etching in c-direction and thus limiting the growth speed in an H-rich environment, whereas in N-polar nanorods the r-facet is Ga-terminated and the c-facet is H-passivated. This leads to an etching of the r-facet and forming a stable c-facet for N-polar rods. In conclusion, this could lead to a higher growth rate of N-polar regimes in an H-rich environment. The H₂:N₂-ratio was raised from 1:33 to 1:10 and the resulting nanorods have been investigated. Based on our results we suggest a dominance of N-polar regimes in dependence of the rod-height, so that after a certain growth time the nanorods become N-polar. As a side effect, the higher amount of H₂ leads to an improvement of morphology due to etching effects on the flanks resulting in regular six-folded nanorods.

As a second objective, the density of nanorods had to be raised. This could be achieved by increasing the V/III ratio. Several ratios have been tested, yielding in the best results for a V/III-ratio of 35 with a wire-density of 39,000 mm⁻². Higher ratios lead to a decrease in uniformity; lower ratios produce far lower numbers. These nanorods show a high homogeneity with standard deviations of 0.62 µm and 0.23 µm in diameter and height respectively. It has been shown that their height can be adjusted from 5 µm to 10 µm without further lateral growth, providing aspect ratios > 6.5.

Silicon is used as n-dopant for GaN. However, Si-atoms lead to the formation of a Silicon-rich layer (most likely SiNₓ) on the surface of MOVPE-grown GaN [TES14], which deteriorate the growth of InGaN. To bypass this effect, an additional n-GaN-shell is introduced between core and InGaN. It has been found that the H₂-concentration in the carrier gas has a crucial influence on the shell growth, reducing the shell height and diameter for higher concentrations. This is due to the formation of Ga-H complexes reducing the diffusion
length of Ga-adatoms on the wire surface. Ga-precursor-decomposition takes place on the wire tip so that reducing the diffusion-length automatically leads to reducing the shell length. The optimal H$_2$-N$_2$-ratio for shell growth has been found to be 1:69 leading to long shells with a low number of defects (Fig. 2).

Fig.2  a) GaN-nanorods with n-GaN-shell. These nanorods show six-fold symmetry, few defects and high homogeneity. b) linear dependency of shell-thickness and shell-height from H$_2$-flow.

On these shell grown InGaN has been analysed by µ-PL and EDX. It could be found that no layer growth took place but InGaN grew selectively on the shell-ends, favouring r-facets. The overall InGaN-growth has been very low.

Finally, a new approach for selective area growth was developed. Using nanoimprintlithography (NIL), 25 nm deep holes with a diameter of 500 nm have been structured into Si (111) via dry etching. In this holes, GaN selectively nucleates. To explain this, the following growth model was developed. MOVPE growth on these substrates leads to nucleation in the lower edges of these holes due to surface energy reduction, similar to GaN growth in epitaxial lateral overgrowth (ELOG, Fig. 3) [NAM98]. These nucleation seeds coagulate and height growth starts.

Fig.3  Selective area growth model. a) Ga-atoms diffuse on the surface and nucleation starts in the NIL-processed holes. b) and c) Nucleation seeds coagulate. d) Height growth of the nucleation seeds.
Conclusion

The MOVPE growth of high quality self-organized GaN-nanorods on Si (111) has been heavily improved. By raising the H₂:N₂ carrier gas ratio, preferential N-polar growth could be achieved. Homogeneity and morphology have been improved as a side-effect. The total amount of nanorods could be raised by the factor 5 by increasing the V/III ratio. To improve InGaN-QW-growth and to enlarge the active area, an n-GaN-interlayer between the n-GaN-core and the QW has been introduced. The carrier-gas-dependency of this shell-growth has been demonstrated and it was explained by a simple growth model. The InGaN-growth has been investigated by µ-PL and EDX, leading to the conclusion of preferential growth on the GaN-r-facet. At last, a world-new maskless approach for selective area epitaxy based on simple substrate preparation by NIL and the preferred epitaxial growth on surface-edges has been demonstrated and described.

References


