True-Green Photonic Crystal LED With Semipolar Quantum Wells Based on Embedded GaN Nanostripes: Polarized and Directional Light Emission

Robert A. R. Leute

A bottom-up approach for photonic crystal emitters and LEDs with semipolar quantum wells on large foreign substrates is presented. Structured c-oriented GaN/AlGaN templates are overgrown with GaN nanostripes and semipolar QWs emitting in the true-green spectral region. After embedding, the stripes and the growth mask act as one-dimensional (1D) photonic crystal. Electroluminescence shows bright green emission. The photonic crystal dispersion relation is simulated and confirmed by angle-resolved spectroscopy, structural analysis is performed by transmission electron microscopy.

1. Introduction

Nitride based light emitting diodes (LEDs) are widely used for lighting and display applications. Especially for the latter application, tailored emission properties, like long wavelength, degree of polarization and directionality are of high interest [1]. Nanostructuring methods allow us to influence both epitaxial growth and the optical properties of our material. The selective epitaxy on a nanoscale allows the embedding of semipolar quantum wells (QWs) with their distinct polarization properties [2] within c-oriented layers [3] while submicrometer periodicities of dielectrics create a photonic crystal (PhC) [4,5] and manipulate the light extraction. Reported PhC LEDs based on InGaN/GaN quantum wells [6, 7] typically use a top-down approach, etching holes into the semiconductor material. We employ a patterned dielectric mask in a dual use as both growth mask and PhC.

2. Experimental

All epitaxial growth takes place inside an Aixtron-200/4RF-S HT metal organic vapor phase epitaxy (MOVPE) reactor with standard precursors TMAl, TMGa, TMIn and high purity ammonia. Silane and Cp_2Mg are used for n- and p-doping, respectively. Firstly, c-oriented GaN/AlGaN templates are grown on 2-inch sapphire substrates. An oxygen doped AlN nucleation layer [8] and an *in-situ* SiN_x nanomask [9] are used for defect reduction. An AlGaN layer of 0.4 µm thickness is used as lower waveguide cladding. 20 nm SiO_2 are deposited via plasma enhanced chemical vapor deposition (PECVD) and

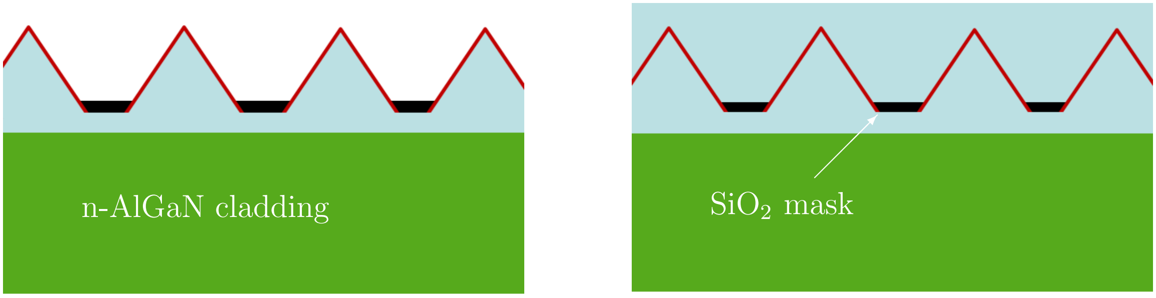

Fig. 1: Schematic drawing of a cross-section of our samples. Nanostripes with semipolar QWs are grown by selective epitaxy (left). After embedding, both QWs and the growth mask acting as 1D PhC are completely buried and the sample has a planar surface (right).

patterned into stripes aligned parallel to GaN a-direction by hot embossing nanoimprint lithography and dry etching with SF_6 plasma. The patterned SiO_2 acts both as 1D PhC and growth mask. GaN nanonstripes are grown by selective epitaxy, including an InGaN prewell. A single QW with 20% indium is deposited on the $\{10\bar{1}1\}$ side facets. After an undoped spacer, the structure is planarized with Mg-doped GaN; exact growth conditions are found in [10]. A schematic of the structure is given in Fig. 1. After growth, the samples are annealed in ambient atmosphere at 750°C for one minute to activate the Mg acceptors. For electrical characterization, 1 µm thick circular indium contacts are evaporated on the surface. Optical output power is measured on-wafer inside an integrating sphere. No additional light extraction methods besides the built-in photonic crystal were used. Angle-resolved photoluminescence (ARPL) spectra are recorded with a TRIAX monochromator and a liquid-nitrogen-cooled charge-coupled device (CCD) at the Institute of Quantum Matter by F. Huber of the Semiconductor Physics Group. The dispersion relation and other modal properties of the PhC are calculated using finite-difference-time-domain (FDTD) simulations [11] using a wavelength dependent dielectric function based on a Lorentz oscillator model; details are found in [12]. The PhC LED structures were investigated by J. Biskupek from the Central Facility of Electron Microscopy via transmission electron microscopy (TEM) after preparation of cross-section TEM samples. Conventional bright- and dark-field TEM experiments for analysis of the crystal structure of the LEDs were carried out using a Philips CM20 operating at 200 kV.

3. Results

3.1 Electroluminescence

The LED shows stable emission, centered at 535 nm, under continuous-wave operation up to 100 mA. Figure 2 shows the power-current-voltage characteristics of the device. Spectra show a broad emission with characteristic modulations, as can be seen in Fig. 3 which will be discussed in subsequent sections.

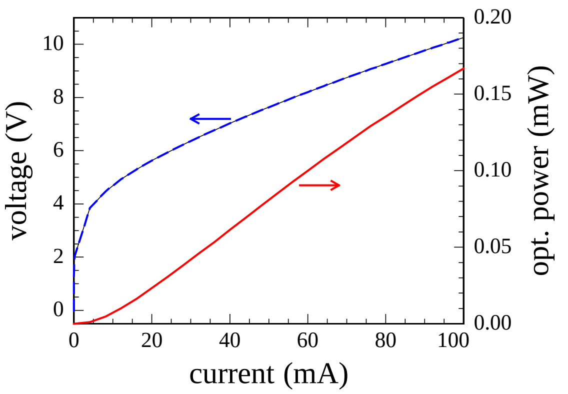

Fig. 2: Voltage-light-current characteristics of the photonic crystal LED.

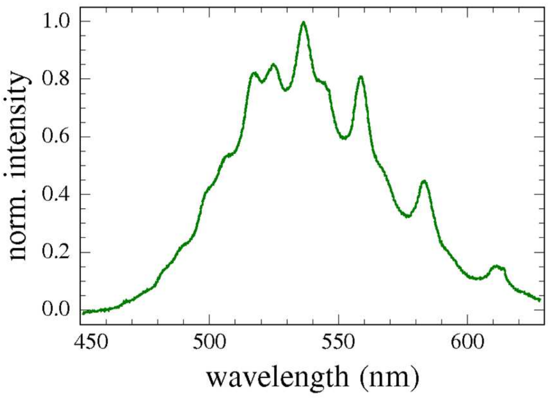

Fig. 3: Electroluminescence spectrum taken at 30 mA. The periodic modulation is caused by the built-in PhC.

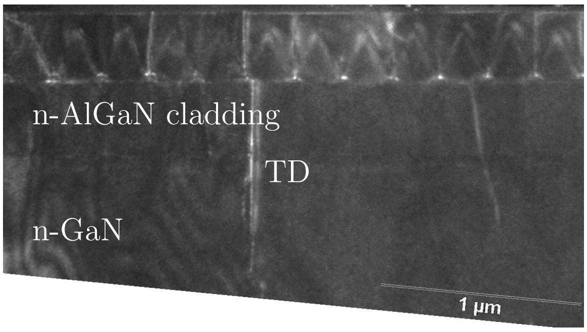

Fig. 4: Weak-beam dark-field TEM image. Few dislocations thread from the sapphire-GaN interface. Some dislocations are created at the coalescence point of the nanostripes.

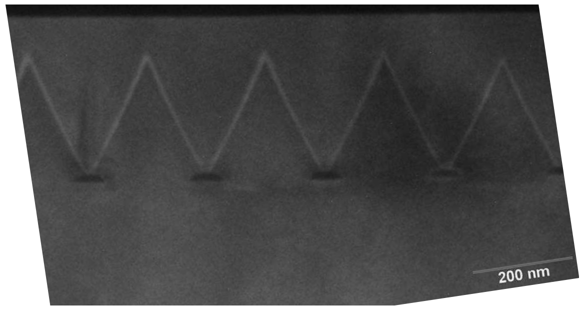

Fig. 5: HACDF image with high Z-contrast. The semipolar QWs show excellent quality and homogeneity despite high indium content.

3.2 Structural analysis

Weak-beam dark-field TEM investigations of our samples show the high quality of the c-oriented heteroepitaxial growth (Fig. 4). Very few dislocations penetrate to the active zone. However, during embedding, new dislocations are formed at the coalescence region, presumably caused by high strain due to the large indium content of the quantum well. In similar samples with QWs emitting in the blue spectral range and lower In content, the active zone maintained the disclocation density of the underlying template [13]. Nevertheless, we find no stacking faults, which are a common challenge to semipolar GaN [14]. The high quality and homogeneity of the QWs, paramount for efficient light emission, relies on a nano-scale precision of all patterning processes and is clearly visible in high-angle center-dark-field (HACDF) images (Fig. 5). The ridge of the nanostripes is as narrow as 2 nm (HRTEM picture not shown).

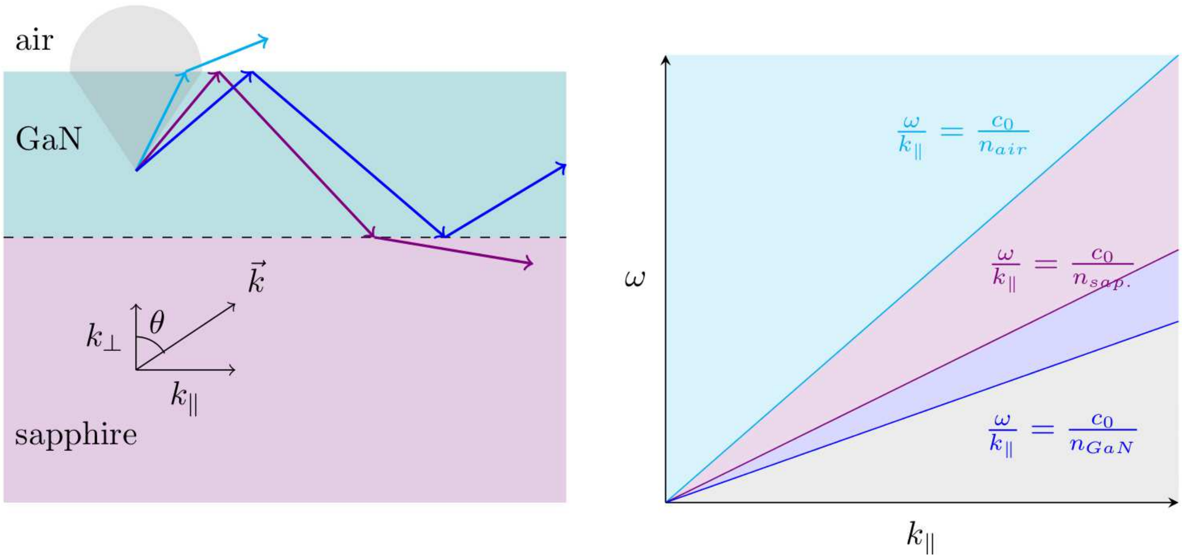

Fig. 6: Waveguiding within a GaN layer on sapphire (left) and the resulting dispersion relation (right). ω is the angular frequency and $k_\parallel$ is the part of the wave vector parallel to the sample surface. In the dispersion relation, the cyan area relates to directly emitted light. Light emitted more shallowly is totally reflected. In the violet area, the light leaks into the substrate. Between the violet sapphire line and the blue GaN line (blue area), the light is guided inside the GaN layer and forms discrete modes. Below the GaN line (gray area) no light propagation is possible.

3.3 Emission properties

Since the embedding of the nanostripes leads to a planar surface of the LED, light emitted by the semipolar QWs will be reflected internally if its angle towards the surface normal θ is larger than the critical angle $\sin\theta_c = n_{air}/n_{GaN}$. If we take the wave vector parallel to the surface $k_\parallel = |\vec{k}|\sin\theta$ with $|\vec{k}| = 2\pi n_{GaN}/\lambda$, the requisition for total reflection is

$$k_{\parallel,c} = |\vec{k}|\sin\theta_c = \frac{2\pi n_{GaN}}{\lambda}\frac{n_{air}}{n_{GaN}}, \quad \omega = 2\pi c/\lambda, n_{air} \approx 1, \quad \Rightarrow k_{\parallel,c} = \frac{\omega}{c}\,. \tag{1}$$

λ is the vacuum wavelength, ω is the angular frequency and c the vacuum velocity of light. The boundary condition of $k_\parallel = \omega/c$ defines the light cone. All emission with k-vectors inside the light cone are emitted directly from the surface. When the k-vector lies outside the light cone, the light is confined within the layer (see Fig. 6).

Discrete modes are formed which store a large part of the energy lost in conventional devices without light extraction mechanisms [15]. The number and spacing of the confined modes depend on the geometry of the device, with the typical thickness of a LED layer stack leading to dozens of modes with narrow linewidths. The presence of a cladding layer below the active zone introduces additional modes which are efficiently pumped (store more energy) due to the large spatial overlap with the emitting QWs (Fig. 7). For non-structured devices, these cladding modes would still be confined and ultimately lost. The periodic variation of the dielectric function caused by the growth mask of the GaN nanostripes acts as photonic crystal. The reciprocal lattice vector $\vec{G}$ of the PhC can be

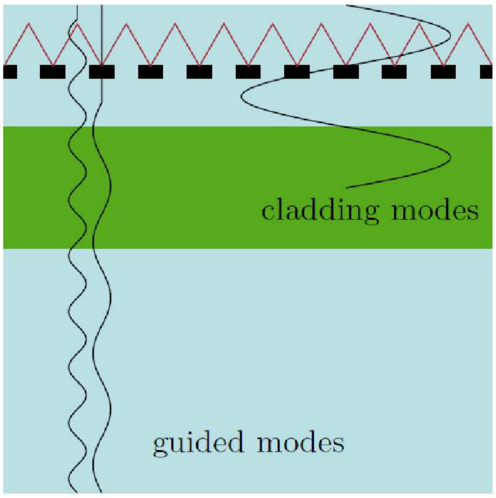

Fig. 7: Introducing an additional cladding layer, most energy is stored inside low order cladding modes with a large overlap with both the QWs and the PhC.

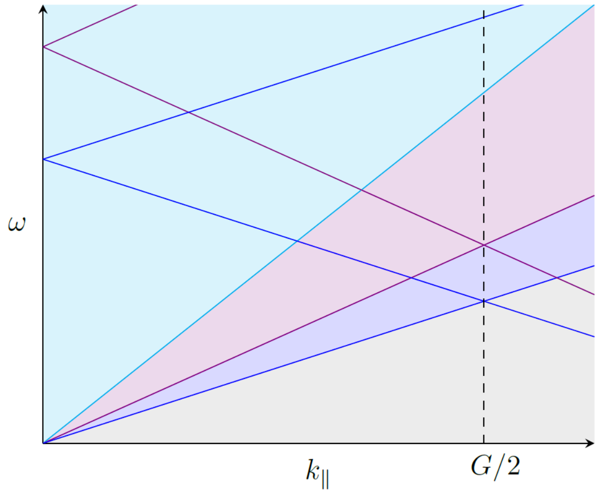

Fig. 8: The PhC leads to a periodicity of the dispersion relation. Confined light (i.e. located between sapphire and GaN line) is folded back into the light cone and extracted.

added to the wave vector of the confined modes $\vec{k}^* = \vec{k} + n \cdot \vec{G}$, folding them back into the light cone and extracting them with high efficiency, shown in Fig. 8. Since the cladding mode stores more energy, than the high-order modes, we obtain a directional emission with a narrow linewidth. Furthermore the semipolar nature of the QWs leads to a high polarization degree [2, 3, 16]. For full analysis of the luminescence properties, the samples are investigated by polarization-dependent ARPL (Fig. 9). Spectral resolution is 0.7 nm and angular resolution is 0.33°.

The flat sample surface leads to typical Fabry-Pérot modulations. The guided modes are extracted by the PhC and appear as sharp resonances. The inclusion of the AlGaN cladding layer leads to a high intensity of few select cladding modes. The very sharp high order modes which propagate in the complete layer stack carry less energy. This preferential selection leads to a strong directionality for the true-green QW emission with highest intensity between 10° and 25°. Comparison of single spectra, taken in this range, show that the cladding mode is highly polarized parallel to the a-direction (parallel to the nanostripes) and exhibits a full-width at half-maximum (FWHM) of 17 meV which gives a Q-factor of 150. The ARPL spectra can be transformed into the dispersion relation (Fig. 10) using the axis transformation (λ, θ) to $(a/\lambda, a/\lambda \cdot \sin(\theta))$ with a being the periodicity of the PhC, which corresponds to reduced frequency and parallel wave vector respectively. The light line marks the border of the light cone. GaN, AlGaN and sapphire line are folded back by the PhC and indicate the area where guided modes can exist. GaN and AlGaN lines are calculated by using a Lorentz oscillator model for the dielectric function [12]. Guided modes within the GaN layer are parallel to the GaN line and confined between GaN line and sapphire line, the cladding mode is localized between GaN and AlGaN line. This is in contrast to the Fabry-Pérot modes which are found without boundary. FDTD simulations with MEEP [11] are in good agreement with the experimental data (not shown).

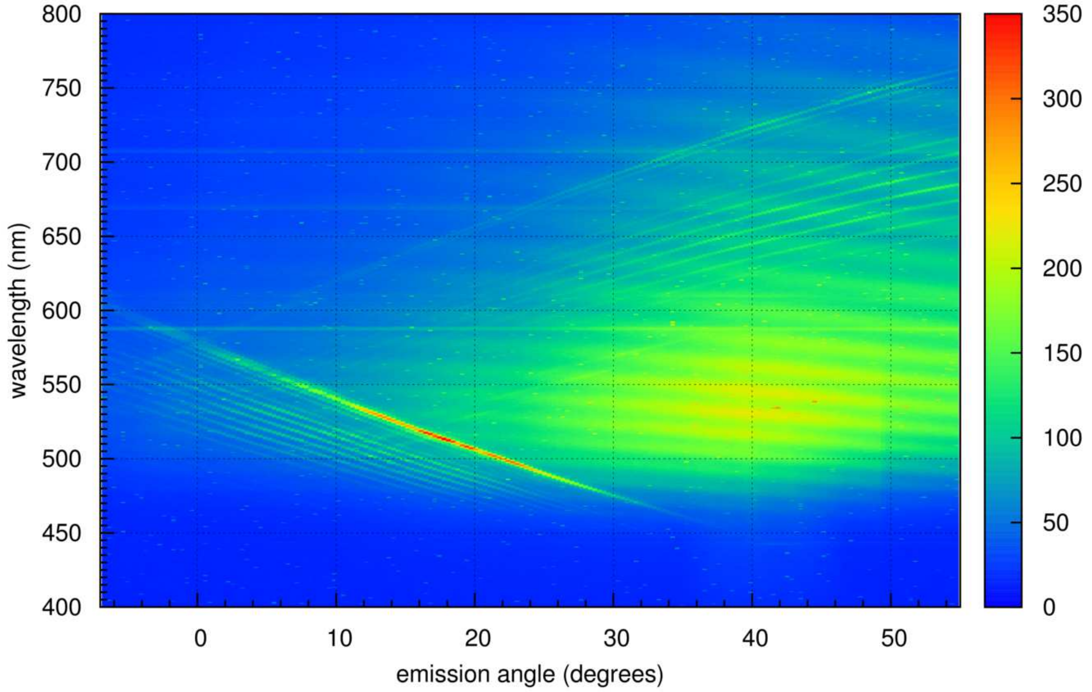

Fig. 9: ARPL spectrum of PhC LED. The polarizer is set parallel to the a-direction. We observe efficient mode outcoupling by the PhC. A single cladding mode has enhanced intensity, is highly polarized and exhibits a narrow linewidth.

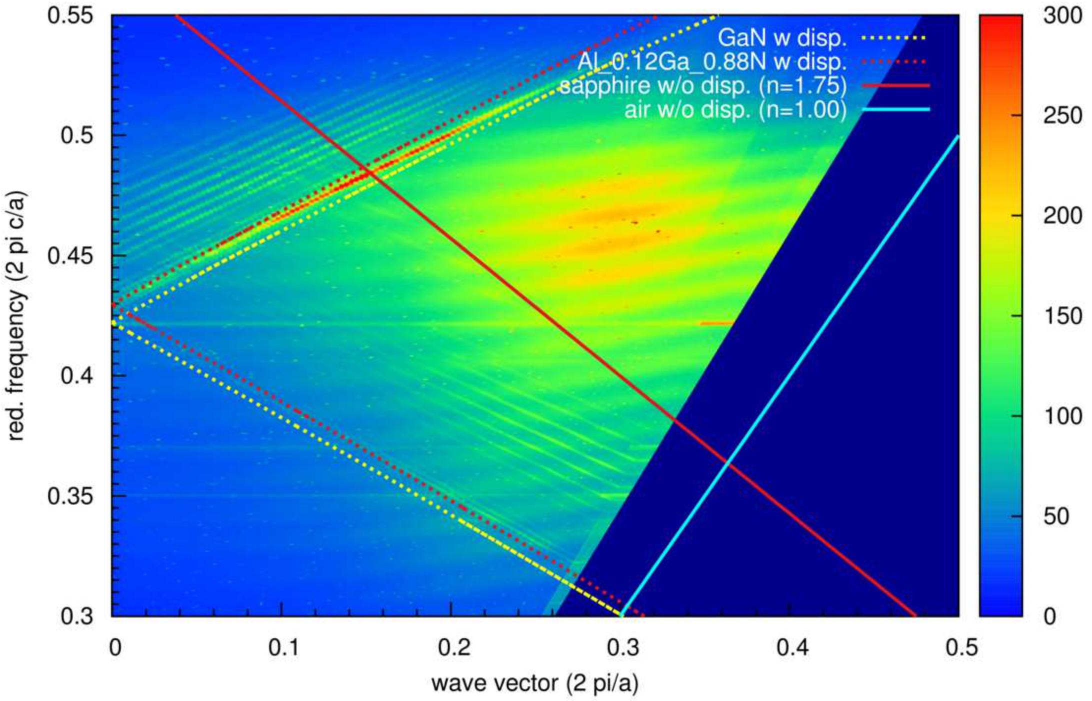

Fig. 10: Dispersion relation of the PhC LED. The cladding mode is localized between the GaN line and AlGaN line.

4. Conclusion

In this work, a PhC LED based on GaN nanostripes with semipolar QWs emitting in the true-green spectral range has been demonstrated on c-sapphire substrates. Nanopatterning was achieved on full 2-inch wafers. TEM analysis shows excellent structural quality, electrical excitation shows stable emission up to 100 mA. The directional extraction of highly polarized cladding modes was demonstrated by angle-resolved photoluminescence.

Acknowledgment

This work was made possible by many fruitful collaborations. Scientific support from D. Heinz, J. Wang, T. Meisch, F. Scholz, F. Huber, K. Thonke, J. Biskupek and U. Kaiser is gratefully acknowledged. Furthermore, I thank J. Harming and C. Thanner from EVG for nanoimprint patterning, R. Rösch and R. Blood for their technical support, B. Neuschl and O. Rettig for fruitful discussions and S. Grözinger for TEM sample preparation. The research was funded by Deutsche Forschungsgemeinschaft (DFG) within the research group FOR957 PolarCoN.

References

[1] C. Wiesmann, K. Bergenek, N. Linder, and U.T. Schwarz, "Photonic crystal LEDs – designing light extraction", *Laser and Photonics Reviews*, vol. 3, pp. 262–286, 2009.

[2] 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.

[3] 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*, vol. 370, pp. 101–104, 2013.

[4] J.D. Joannopoulos, P.R. Villeneuve, and S. Fan, "Guided modes in photonic crystal slabs", *Nature*, vol. 386, pp. 143–149, 1997.

[5] D. Heinz, "Zweidimensionale photonische Kristalle basierend auf nitridischen Halbleitern", Diplomarbeit, Universität Ulm, Institut für Quantenmaterie – Gruppe Halbleiterphysik der Fakultät für Naturwissenschaften, 2011.

[6] K. McGroddy, A. David, E. Matioli, M. Iza, S. Nakamura, S. DenBaars, J.S. Speck, C. Weisbuch, and E.L. Hu, "Directional emission control and increased light extraction in GaN photonic crystal light emitting diodes", *Appl. Phys. Lett.*, vol. 93, pp. 103502-1–3, 2008.

[7] E. Matioli and C. Weisbuch, "Impact of photonic crystals on LED light extraction efficiency: approaches and limits to vertical structure designs", vol. 43, pp. 354005-1–6, 2010.

[8] B. Kuhn and F. Scholz, "An oxygen doped nucleation layer for the growth of high optical quality GaN on sapphire", *Phys. Status Solidi A*, vol. 188, pp. 629–633, 2001.

[9] 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.

[10] R.A.R. Leute, J. Wang, T. Meisch, J. Biskupek, U. Kaiser, and F. Scholz, "Blue to true green LEDs with semipolar quantum wells based on GaN nanostripes", *Phys. Status Solidi C*, 2014, DOI: 10.1002/pssc.201400176.

[11] A.F. Oskooi, D. Roundy, M. Ibanescu, P. Bermel, J.D. Joannopoulos, and S.G. Johnson, "MEEP: A flexible free-software package for electromagnetic simulations by the FDTD method", *Comput. Phys. Commun.*, vol. 181, pp. 687–702, 2010.

[12] D. Heinz, R.A.R. Leute, S. Kizir, Y. Li, T. Meisch, K. Thonke, and F. Scholz, "Ga(In)N photonic crystal light emitters with semipolar quantum wells", *Jpn. J. Appl. Phys.*, vol. 52, pp. 062101-1–5, 2013.

[13] R.A.R. Leute, T. Meisch, J. Wang, J. Biskupek, U. Kaiser, M. Muller, P. Veit, F. Bertram, J. Christen, and F. Scholz, "GaN laser structure with semipolar quantum wells and embedded nanostripes", in Proc. *10th Conf. on Lasers and Electro-Optics Pacific Rim (CLEO-PR)*, paper WH3-4, 2 pages, June 2013.

[14] F. Wu, Y. Zhao, A. Romanov, S.P. DenBaars, S. Nakamura, and J.S. Speck, "Stacking faults and interface roughening in semipolar $(20\bar{2}\bar{1})$ single InGaN quantum wells for long wavelength emission", *Appl. Phys. Lett.*, vol. 104, pp. 151901-1–5, 2014.

[15] T. Fujii, Y. Gao, R. Sharma, E.L. Hu, S.P. DenBaars, and S. Nakamura, "Increase in the extraction efficiency of GaN-based light-emitting diodes via surface roughening", *Appl. Phys. Lett.*, vol. 84, pp. 855–857, 2004.

[16] Y. Zhao, R.M. Farrell, Y.R. Wu, and J.S. Speck, "Valence band states and polarized optical emission from nonpolar and semipolar III-nitride quantum well optoelectronic devices", *Jpn. J. Appl. Phys.*, vol. 53, pp. 100206-1–17, 2014.