Heteroepitaxial Growth of Planar Semipolar $(10\bar{1}1)$ GaN by Metalorganic Vapor Phase Epitaxy

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We report on the growth of planar semipolar $(10\bar{1}1)$ *GaN on* $(11\bar{2}3)$ *pre-patterned sapphire. This method allows the growth of semipolar oriented* $(10\bar{1}1)$ *GaN on large scale. By x-ray diffaction, only the peaks of the desired* $(10\bar{1}1)$ *plane could be observed. The in-plane orientations could be determined as* $[0001]_{GaN} \parallel [0001]_{sapphire}$ *and* $[11\bar{2}0]_{GaN} \parallel [10\bar{1}0]_{sapphire}$. *Scanning electron microscopy, transmission electron microscopy and atomic force microscopy measurements show coalesced surfaces with root mean square roughness values below 2 nm (10 µm×10 µm). Further investigations using photoluminescence spectroscopy show spectra that are dominated by the near band edge emission. The high crystal quality is confirmed by small full width at half maximum values of x-ray rocking curve measurements of less than 400 arcsec.*

1. Introduction

Devices like light emitting diodes (LEDs) based on GaN are usually grown in c-direction. Due to induced biaxial strain and the lattice geometry of group-III nitrides, huge piezoelectric fields are present within heterostructures along this particular direction. The resulting band bending causes some undesirable effects on the quantum wells (QWs) grown in that direction [1], like spatial separation of the wave functions of electrons and holes. Consequently, the recombination probability (recombination rate) of electrons and holes is reduced, the emission wavelength is redshifted and dependent on the drive current due to screening [2]. This is known as quantum confined Stark effect (QCSE).

One possibility to reduce these negative effects is to grow in semipolar or nonpolar direction. Due to the lack of real bulk GaN substrates, nowadays these structures have to be grown on foreign substrates and therefore the research is still focussed on the growth on different templates. Nonpolar GaN can be grown on several different foreign substrates but still suffers from a huge amount of stacking faults [3–5]. Various semipolar orientations of GaN have been also investigated on different substrates, like $(10\bar{1}1)$ GaN on silicon [6], $MgAl_2O_4$ [7], or as facets grown on c-plane sapphire [8]. $(11\bar{2}2)$ GaN has been grown on m-plane sapphire [9] or on facets of c-plane oriented GaN stripes [10]. Just recently Okada et al. presented a method to grow $(11\bar{2}2)$ GaN on r-plane sapphire [11]. Another possible approach is to use sliced pieces from hydride vapor phase epitaxial (HVPE) grown material [12, 13], but these templates are quite expensive and very limited in size. Up to now the perfect substrate is still missing and therefore there is a lot of research in this field at the moment.

From this background, in this study, we propose the planar growth of semipolar $(10\bar{1}1)$ oriented GaN directly on pre-patterned sapphire (Fig. 1) in a similar approach as Hikosaka et al. or Okada et al. [6, 11]. For this purpose the use of $(11\bar{2}3)$ sapphire seems to be

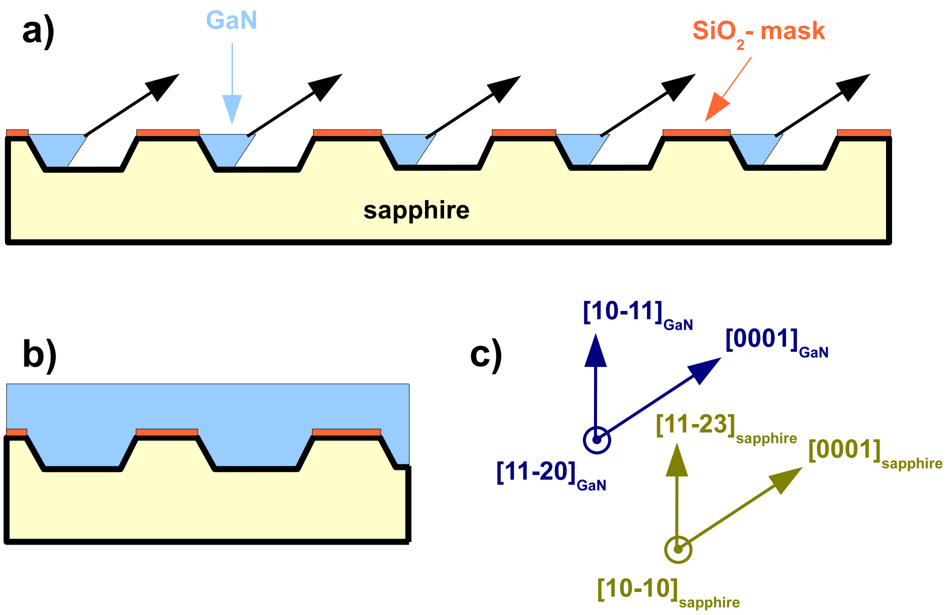

Fig. 1: Schematic figure of the basic idea. a) Grooves are etched into $(11\bar{2}3)$ sapphire, the growth starts at the sidewalls in c-direction. b) The GaN islands coalesce, resulting in a planar semipolar $(10\bar{1}1)$ GaN layer. c) The crystal orientations of the sapphire and the semipolar GaN.

appropriate as the inclination angle between $(0001)_{\mathrm{Al_2O_3}}$ and $(11\bar{2}3)_{\mathrm{Al_2O_3}}$ is nearly the same like the angle between $(0001)_{\mathrm{GaN}}$ and $(10\bar{1}1)_{\mathrm{GaN}}$. Calculations show that the piezoelectric fields within QWs grown in that direction will be drastically reduced when compared to c-plane growth [14]. Additionally, this surface is regarded as naturally stable facet since it exhibits an automatically formed and very smooth surface [15]. A higher indium incorporation efficiency is also observed, which can be advantageous for longer wavelength light emitters [16].

2. Experimental

As starting substrate n-oriented sapphire was used. The $(11\bar{2}3)$-surface was lithographically structured with grooves along the in-plane m-direction. Therefore we deposited a 200 nm silicon dioxide (SiO_2) mask via plasma-enhanced chemical vapor deposition which also acts as a mask for the selective area growth (SAG). A 550 nm thick mask of nickel and gold, structured with a (3 µm opening) x (3 µm mask) stripe pattern was used for dry etching of the sapphire via reactive ion etching (RIE). After removal of the remaining mask material, the sapphire exhibits approximately 1.2 µm deep grooves, each with one smooth c-plane-like side facet. The subsequent growth was carried out in a commercial horizontal flow Aixtron-200/4 RF-S HT reactor with the standard precursors trimethylgallium (TMGa), trimethylaluminum (TMAl) and high purity ammonia (NH_3). Pd diffused hydrogen was used as carrier gas. The process temperature was controlled by a pyrometer at the backside of the rotation tray. To start growth we used an oxygen doped low temperature AlN nucleation layer [17, 18], followed by approximately 1 µm GaN with a V/III ratio of 650 at a temperature of 1130°C and a pressure of 150 hPa, resulting in stripes of semipolar GaN. Simply elongating the growth time leads to coalesced layers. The growth rate was roughly 2.2 µm/h (on closed layers).

To characterize the samples and their crystal quality we used x-ray diffraction (XRD) rocking curve measurements (XRC) and ω-2θ scans as well as low temperature (14 K) photoluminescence (PL) measurements. Particularly, the latter enables to judge about typical defects in semi- and nonpolar GaN layers, like basal plane stacking faults (BSFs) [19]. The Surface quality could be accessed via scanning electron microscopy (SEM), transmission electron microscopy (TEM), optical phase contrast microscopy (OM) and atomic force microscopy (AFM).

3. Results and Discussion

Figure 1 shows the growth principle. The GaN growth starts from the groove facets of the sapphire wafer in the usual c-direction, which has a certain inclination to the surface (Fig. 1 a) resulting in a flat and planar semipolar layer (Fig. 1 b). The crystal orientation was measured via a symmetrical XRD ω-2θ-scan (Fig. 2). The sapphire substrate peaks

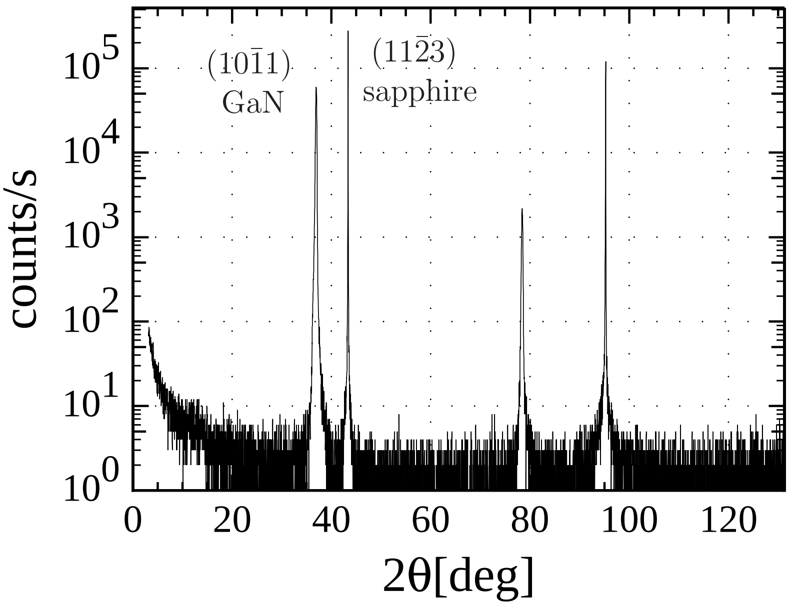

Fig. 2: XRD ω-2θ scan showing the peaks of $(11\bar{2}3)$ sapphire and $(10\bar{1}1)$ GaN (plus second order peaks). No other peak owing to another orientation of GaN is visible.

and the $(10\bar{1}1)$-reflection of GaN are clearly visible; no other crystal orientation could be observed. Additionally, the skew geometric (0002) peak of GaN and the (0006) reflection of sapphire appear at the same φ-angle (rotation around the surface normal) and at the expected χ-angle (rotation around the cut of the surface with the scattering plane)[not shown]. Therefore we conclude an orientation of the GaN film as sketched in Fig. 1 with a semipolar $(10\bar{1}1)$ surface. The in-plane orientations were investigated by selected area electron diffraction of cross-section samples and were found to be $[0001]_{\mathrm{GaN}} \parallel [0001]_{\mathrm{sapphire}}$ and $[11\bar{2}0]_{\mathrm{GaN}} \parallel [10\bar{1}0]_{\mathrm{sapphire}}$.

The SEM micrograph (Fig. 3) reveals the morphology of the samples. The GaN starts to

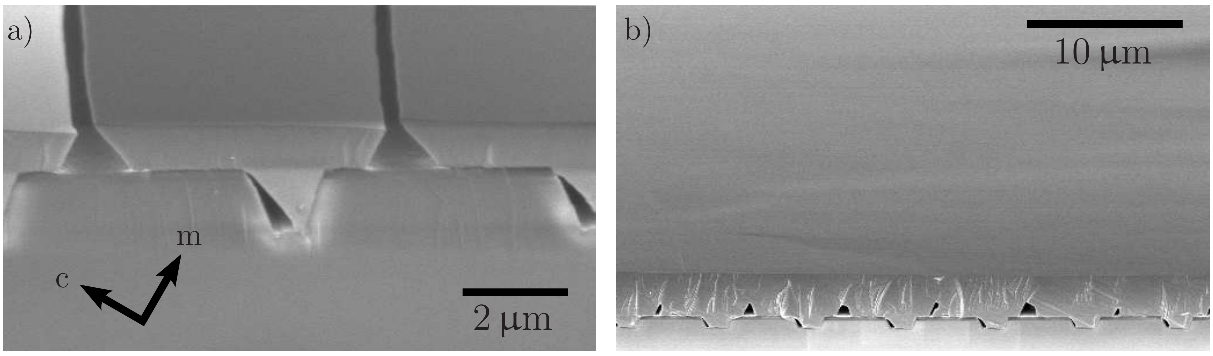

Fig. 3: a) SEM micrograph of the cross-section of an uncoalesced sample. The crystal orientations are the same as sketched in fig 1c). b) Coalesced layer with flat surface.

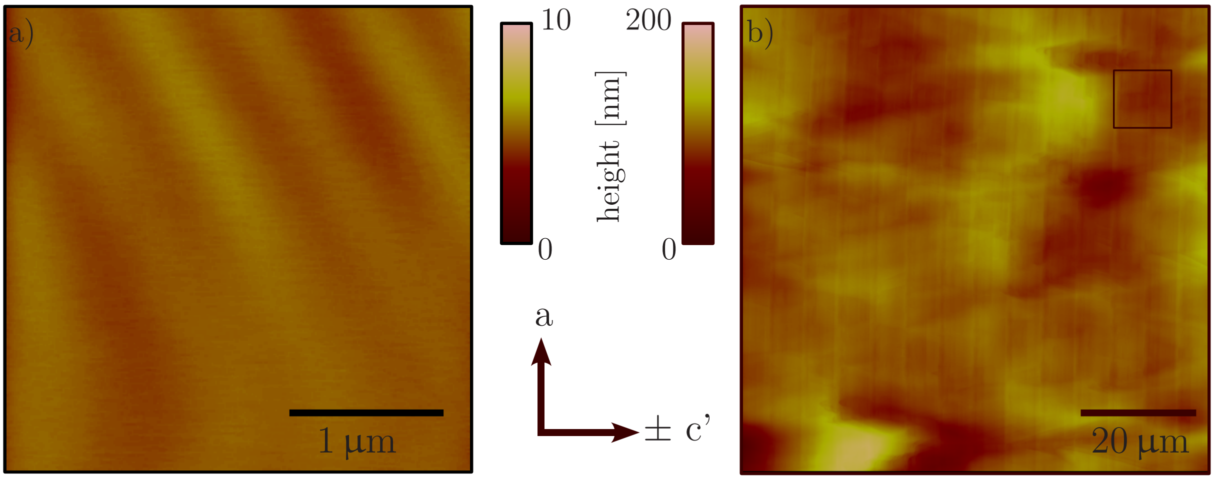

Fig. 4: Surface morphology of $(10\bar{1}1)$-oriented stripes (a) and of an coalesced layer (b). The root mean square roughness (rms) values have measured to approx. 0,2 nm (a, 3 µm × 3 µm measurement) and to approx. 15 nm (b, 80 µm × 80 µm measurement). A 10 µm × 10 µm area from b) results in 1 nm.

grow within the grooves of the prepared sapphire substrate. As proposed, this happens only on the c-plane like side-facet of the trench, resulting in growth directed exclusively in c-directon. Although a complete nucleation layer was deposited, no growth took place on the SiO_2 covered ridges. This area could be overgrown comparable to the well known epitaxial lateral overgrowth (ELOG) principle when the GaN reaches the height of the trenches and is able to grow more in lateral direction. The wings in the two different directions show different behavior. The so called "+c"-wing growing directly in "+c"-direction exhibits a semipolar $(10\bar{1}1)$ facet, whereas the slower grown "-c"-wing is terminated by a "-c"-facet. The wing tilt of the overgrown area is about 0.2° ("+c"-wing), as visible in XRD measurements.

By elongating the growth time coalescence of the stripes could be achieved. We assume that this takes place similar as for other orientations [20]: The fast developing "+c"-wing buries the "-c"-wing and leads to planar and flat surfaces. The excellent surface quality was confirmed by AFM and TEM measurements (Fig. 4). The root mean square roughness determined by AFM was as small as 0.1 nm for a 1 μm×1 μm-scan and below 0.3 nm for a 3 μm×3 μm-scan, respectively. An atomically flat surface was found by HR-TEM investigations [21].

The high crystal quality was verified by narrow XRC peaks. The full width at half maximum (FWHM) of both, the symmetrical $(10\bar{1}1)$ reflection and the asymmetrical (0002) and $(10\bar{1}2)$ reflections were smaller than 400 arcsec, respectively. Furthermore, the low-temperature PL-spectra (Fig. 5) revealed a comparably strong and dominating near band edge emission (NBE) at 3.464 eV (slightly strained) for the stripes and at 3.471 eV (nearly unstrained) for the coalesced sample. Typically, in semi- and nonpolar GaN

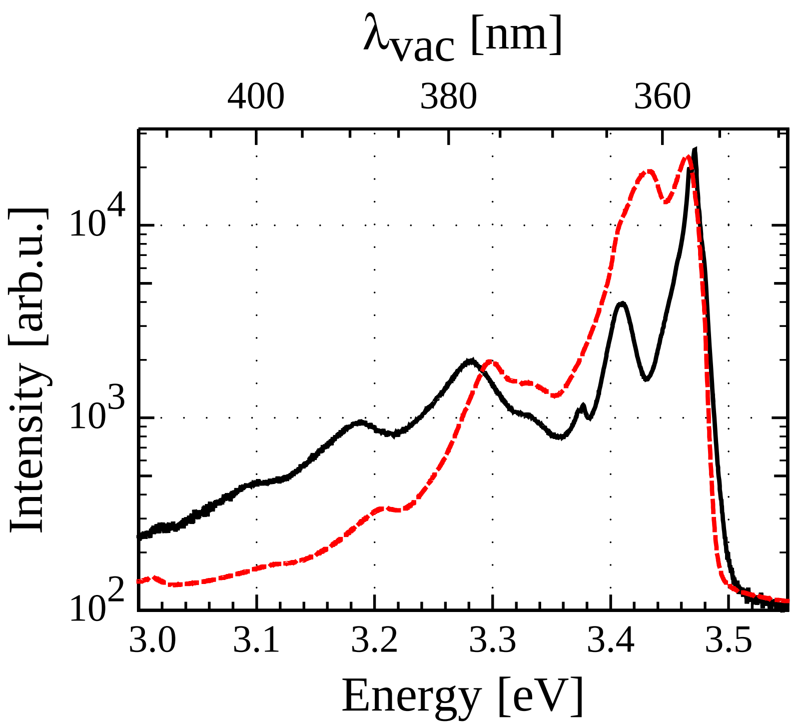

Fig. 5: Photoluminescence spectra recorded at low temperature (15 K) for uncoalesced (dashed) and coalesced (solid line) samples. Visible are the NBE as dominant peak (3.464 eV for stripes, 3.471 eV for the coalesced sample) and some defect related emission lines (e.g. 3.430 eV or 3.300 eV).

grown on unpatterned sapphire this luminescence is quite weak and the defect correlated luminescence is dominating. Nevertheless some of the typical defect related peaks are also visible in our samples. The transition around 3.41 eV, which can be attributed to basal plane stacking faults [22], could not be suppressed completely. Also, the lower energy peaks (around 3.30 eV), usually assigned to (pyramidal) stacking faults (and partial dislocations) [23] are still visible. The local distribution of stacking faults was investigated by transmission electron microscopy (Fig. 6). The highest density of stacking faults can

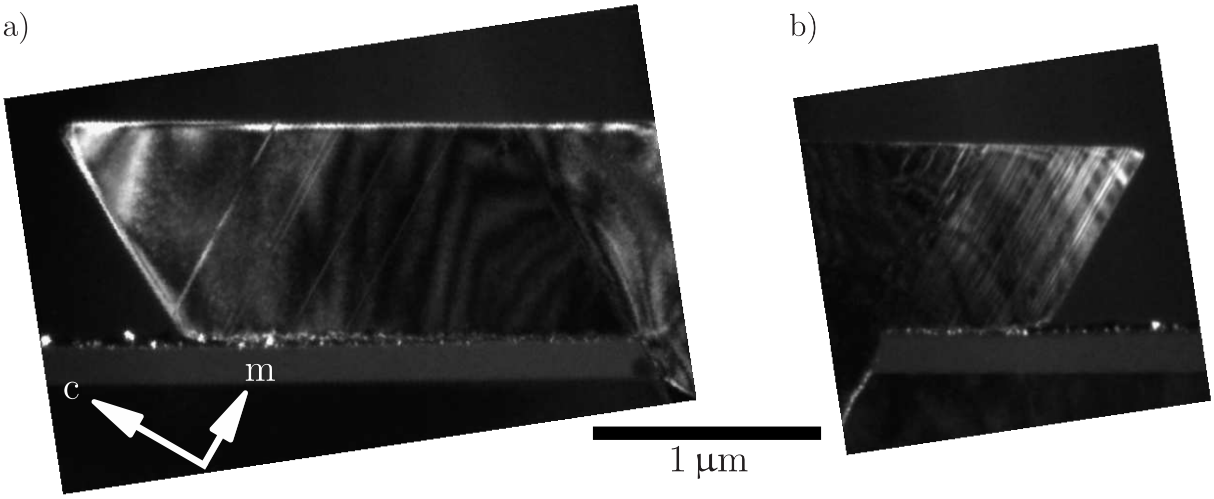

Fig. 6: TEM micrograph of "+c"-wing (a) and "-c"-wing (b) of an uncoalesced sample. (Basal plane) stacking faults are visible as white lines running perpendicular to the c-direction.

be found in the "-c"-wing, the "+c"-wing of the laterally overgrown area provides the lowest density.

4. Conclusion

In summary, planar semipolar $(10\bar{1}1)$ GaN on $(11\bar{2}3)$ pre-patterned sapphire was successfully grown. This method allows large area growth of semipolar oriented $(10\bar{1}1)$ GaN on sapphire. Compared to other growth techniques and the resulting quality of nonpolar GaN on sapphire this approach is quite promising, in particular if further optimization steps are included.

5. Acknowledgement

The author likes to thank Jürgen Däubler for the measurements and results during his diploma thesis and Johannes Biskupek (Central Facility of Electron Microscopy, University of Ulm) for transmission electron microscopy measurements. Additionally the support of Rudolf Rösch, Ilona Argut and Rainer Blood ist acknowledged. This work was financially supported by the Deutsche Forschungsgemeinschaft within the project "Polarization Field Control in Nitride Light Emitters (PolarCoN)" under contract no. Scho 393/22–1.

References

[1] F. Bernardini, V. Fiorentini, and D. Vanderbilt, "Spontaneous polarization and piezoelectric constants of III-V nitrides", *Phys. Rev. B*, vol. 56, pp. R10024–R10027, 1997.

[2] T. Takeuchi, C. Wetzel, S. Yamaguchi, H. Sakai, H. Amano, and I. Akasaki, "Determination of piezoelectric fields in strained GaInN quantum wells using the quantum-confined stark effect", *Appl. Phys. Lett.*, vol. 73, pp. 1691–1693, 1998.

[3] M.D. Craven, S.H. Lim, F. Wu, J.S. Speck, and S.P. DenBaars, "Structural characterization of nonpolar $(11\bar{2}0)$ a-plane GaN thin films grown on $(1\bar{1}02)$ r-plane sapphire", *Appl. Phys. Lett.*, vol. 81, pp. 469–471, 2002.

[4] B. Imer, F. Wu, M.D. Craven, J.S. Speck, and S.P. DenBaars, "Stability of $(1\bar{1}00)$ m-plane GaN films grown by metalorganic chemical vapor deposition", *Jpn. J. Appl. Phys.*, vol. 45, pp. 8644–8647, 2006.

[5] A-T. Cheng, Y-K. Su, W-C. Lai, and Y-Z. Chen, "Metalorganic vapor phase epitaxy growth of m-plane GaN using $LiAlO_2$ substrates", *Jpn. J. Appl. Phys.*, vol. 47, pp. 3074–3076, 2008.

[6] T. Hikosaka, T. Narita, Y. Honda, M. Yamaguchi, and N. Sawaki, "Optical and electrical properties of (1-101) GaN grown on a 7° off-axis (001) Si substrate", *Appl. Phys. Lett.*, vol. 84, pp. 4717–4719, 2004.

[7] T.J. Baker, B.A. Haskell, F. Wu, P.T. Fini, J.S. Speck, and S. Nakamura, "Characterization of planar semipolar gallium nitride films on spinel substrates", *Jpn. J. Appl. Phys.*, vol. 44, pp. L920–L922, 2005.

[8] T. Wunderer, P. Brückner, B. Neubert, F. Scholz, M. Feneberg, F. Lipski, M. Schirra, and K. Thonke, "Bright semipolar GaInN/GaN blue light emitting diode on side facets of selectively grown GaN stripes", *Appl. Phys. Lett.*, vol. 89, pp. 041121-1–3, 2006.

[9] T.J. Baker, B.A. Haskell, F. Wu, J.S. Speck, and S. Nakamura, "Characterization of planar semipolar gallium nitride films on sapphire substrates", *Jpn. J. Appl. Phys.*, vol. 45, pp. L154–L157, 2006.

[10] K. Nishizuka, M. Funato, Y. Kawakami, S. Fujita, Y. Narukawa and T. Mukai, "Efficient radiative recombination from $(11\bar{2}2)$-oriented $In_xGa_{1-x}N$ multiple quantum wells fabricated by the regrowth technique", *Appl. Phys. Lett.*, vol. 85, 3122–3124, 2004.

[11] N. Okada, A. Kurisu, K. Murakami, and K. Tadatomo, "Growth of semipolar $(11\bar{2}2)$ GaN layer by controlling anisotropic growth rates in r-plane patterned sapphire substrate", *Appl. Phys. Exp.*, vol. 2, pp. 091001-1–3, 2009.

[12] M. Ueda, K. Kojima, M. Funato, Y. Kawakami, Y. Narukawa, and T. Mukai, "Epitaxial growth and optical properties of semipolar $(11\bar{2}2)$ GaN and InGaN/GaN quantum wells on GaN bulk substrates", *Appl. Phys. Lett.*, vol. 89, pp. 211907-1–3, 2006.

[13] T. Paskova, R. Kroeger, S. Figge, D. Hommel, V. Darakchieva, B. Monemar, E. Preble, A. Hanser, N.M. Williams, and M. Tutor, "High-quality bulk a-plane GaN sliced from boules in comparison to heteroepitaxially grown thick films on r-plane sapphire", *Appl. Phys. Lett.*, vol. 89, pp. 051914-1–3, 2006.

[14] M. Feneberg, F. Lipski, R. Sauer, K. Thonke, T. Wunderer, B. Neubert, P. Brückner, and F. Scholz, "Piezoelectric fields in GaInN/GaN quantum wells on different crystal facets", *Appl. Phys. Lett.*, vol. 89, pp. 242112-1–3, 2006.

[15] T. Wunderer, F. Lipski, J. Hertkorn, P.Brückner, F. Scholz, M. Feneberg, M. Schirra, K. Thonke, A. Chuvilin, and U. Kaiser, "Bluish-green semipolar GaInN/GaN light emitting diodes on $1\bar{1}01$ GaN side facets", *phys. stat. sol. (c)*, vol. 5, pp. 2059–2062, 2008.

[16] F. Scholz, T. Wunderer, B. Neubert, M. Feneberg, and K. Thonke, "GaN-based light-emitting diodes on selectively grown semipolar facets", *MRS Bull.*, vol. 34, pp. 328–333, 2009.

[17] B. Kuhn, and F. Scholz, "An oxygen doped nucleation layer for the growth of high optical quality GaN on sapphire", *phys. stat. sol. (a)*, vol. 188, pp. 629–633, 2001.

[18] J. Hertkorn, P. Brückner, S.B. Thapa, T. Wunderer, F. Scholz, M. Feneberg, K. Thonke, R. Sauer, M. Beer, and J. Zweck, "Optimization of nucleation and buffer layer growth for improved GaN quality", *J. Crystal Growth*, vol. 308, pp. 30–36, 2007.

[19] R. Liu, A. Bell, F.A. Ponce, C.Q. Chen, J.W. Wang, and M.A. Khan, "Luminescence from stacking faults in gallium nitride", *Appl. Phys. Lett.*, vol. 86, pp. 021908-1–3, 2005.

[20] S. Schwaiger, S. Metzner, T. Wunderer, I. Argut, J. Thalmair, F. Lipski, M. Wieneke, J. Bläsing, F. Bertram, J. Zweck, A. Krost, J. Christen, and F. Scholz, "Growth and coalescence behavior of semipolar $(11\bar{2}2)$ GaN on pre-structured r-plane sapphire substrates", *phys. stat. sol. (b)*, DOI:10.1002/pssb.201046336 (6 pages).

[21] S. Schwaiger, I. Argut, T. Wunderer, R. Rösch, F. Lipski, J. Biskupek, U. Kaiser, and F. Scholz, "Planar semipolar $(10\bar{1}1)$ GaN on $(11\bar{2}3)$ sapphire", *Appl. Phys. Lett.*, vol. 96, pp. 231905-1–3, 2010.

[22] T. Paskova (Ed.), *Nitrides with Nonpolar Surfaces. Growth, Properties and Devices*, Weinheim: Wiley-VCH Verlag, 2008.

[23] P.P. Paskov, R. Schifano, B. Monemar, T. Paskova, S. Figge, and D. Hommel, "Emission properties of a-plane GaN grown by metalorganic chemical-vapor deposition", *J. Appl. Phys.*, vol. 98, pp. 093519-1–7, 2005.