

Efficient 350 nm LEDs on low edge threading dislocation density AlGa_N buffer layers

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ABSTRACT

Improving the crystal quality of AlGa_N epitaxial layers is essential for the realization of efficient III-nitride-based light emitting diodes (LEDs) with emission wavelengths below 365 nm. Here, we report on two different approaches to improve the material quality of AlGa_N buffer layers for such UV-LEDs, which are known to be effective for the MOVPE growth of GaN layers. Firstly, we grew AlGa_N on thin GaN nucleation islands which exhibit a three-dimensional faceted structure (3D GaN nucleation). Lateral overgrowth of these islands results in a lateral bending of dislocation lines at the growing facets. Secondly, in-situ deposited SiN_x interlayers have been used as nano-masks reducing the dislocation density above the SiN_x layers. Both approaches result in reduced asymmetric HRXRD ω -scan peak widths, indicating a reduced edge-type dislocation density. They can be applied to the growth of AlGa_N layers with an Al concentration of at least 20%, thus suitable for LEDs emitting around 350 nm. On-wafer electroluminescence measurements at 20 mA show an increase in output power by a factor of 7 and 25 for LED structures grown on 3D GaN nucleation and SiN_x interlayer, respectively, compared to structures grown on a purely 2D grown low Al-content AlGa_N nucleation layer. Mesa-LEDs fabricated from the LED layer sequences grown on buffers with SiN_x interlayer exhibit a low forward voltage of 3.8 V at 20 mA and a maximum continuous wave (cw) output power of 12.2 mW at 300 mA.

Keywords: LED, ultraviolet, AlGa_N, MOVPE, dislocation

1. INTRODUCTION

Light-emitting diodes (LEDs) based on AlGa_N allow to cover emission wavelengths below 365 nm, and are thus solid-state light sources suitable for several applications, e.g. industrial curing, disinfection or fluorescence spectroscopy. However, the efficiency of such LEDs is still about one order of magnitude lower compared GaN-based LEDs emitting at wavelengths above 365 nm [1]. The key limiting factor is the high density of threading edge-type dislocations in AlGa_N buffer layers which penetrate into the active region and act as non-radiative recombination centers [2]. Hence, an improvement in the crystal quality of AlGa_N buffer layers is mandatory for achieving higher internal quantum efficiencies and thus output powers for LEDs emitting below 365 nm [3].

In this work, we demonstrate 350-nm-LEDs grown on AlGa_N with reduced defect density using two different approaches which are known to be effective for MOVPE-grown GaN layers: The first one is growth on three-dimensional GaN nucleation islands leading to a lateral bending of dislocation lines at the growing facets during overgrowth [4]. The second one is the in-situ deposition and overgrowth of SiN_x interlayers acting as nano-masks and reducing the dislocation density above the interlayers [5]. Both approaches have recently been adapted to AlGa_N with Al contents of up to 20 % [6,7]. A reduction of the threading edge-type dislocations is inferred from a decreased peak width in high-resolution X-ray diffraction (HRXRD) ω -scans. An appropriate model for the effect of SiN_x interlayers on the density of edge-type dislocations has been developed based on a transmission electron microscopy (TEM) analysis [8]. The beneficial influence of the reduced defect density on the quantum efficiency of 350-nm-LEDs has already been demonstrated using excitation-density and temperature dependent photoluminescence (PL) measurements [9]. Here, we

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report on the improved electroluminescence performance of these low defect density 350-nm-LEDs measured both on-wafer as well as on fully processed and flip-chip mounted mesa-LEDs.

2. EXPERIMENTAL DETAILS

All samples were grown on 2" c-plane (0001) sapphire substrates by metalorganic vapor phase epitaxy (MOVPE) in an AIXTRON 200/4 RF-S horizontal reactor using standard TMGa, TMAI, SiH₄, Cp₂Mg and NH₃ precursors. The LED structure consists of a 700 nm thick Si-doped Al_{0.15}Ga_{0.85}N n-contact layer and 3 nm thick GaN single quantum well (QW) sandwiched between Al_{0.15}Ga_{0.85}N barriers. On the p-side a 20 nm thick Mg-doped Al_{0.3}Ga_{0.7}N electron blocking layer was grown first, followed by a 50 nm thick Al_{0.15}Ga_{0.85}N:Mg and a 20 nm thick GaN:Mg contact layer. This LED structure was grown on three different AlGaN buffer layers: (1) Al_{0.2}Ga_{0.8}N buffer layer grown on a low-temperature GaN nucleation with a 3D faceted structure, (2) Al_{0.2}Ga_{0.8}N buffer layer grown on an AlN nucleation layer with a sub-monolayer SiN_x mask (this buffer layer has been grown in another similar epitaxial machine [7]), and (3) similar AlGaN buffer layer to (1) with slightly reduced Al content (for the prevention of cracking) grown on a low Al content AlGaN nucleation grown under similar conditions as the GaN nucleation of (1). In contrast to (1), the low Al content of the AlGaN nucleation used in (3) prevents a roughening of the surface during annealing, which results in a purely two-dimensional growth mode. Since buffer layer (3) exhibits no special features for defect reduction, it serves as a benchmark for the other two approaches.

Epitaxial layer growth was monitored by in-situ normal incidence reflectance measurements. Furthermore, the surface morphology at different growth stages was analyzed by ex-situ by atomic force microscopy (AFM). The crystalline quality of the buffer layers was assessed by HRXRD ω -scans using a PANalytical MRD diffraction system equipped with a line focus from a sealed Cu X-ray tube. The ω -scans were performed with a parabolically bent graded multilayer mirror and a two-bounce Ge 220 monochromator at the primary beam side, and a triple axis Ge 220 analyzer set-up at the detector side. AlGaN buffer layers using the SiN_x interlayer approach (3) were analyzed by TEM in weak beam dark field (WBDF) mode on a Philips CM-20 microscope after standard sample preparation including mechanical polishing and low-angle argon beam thinning.

The LED structures were characterized by on-wafer electroluminescence (EL) measurements, using evaporated Ni/Al/Ni/Au p-contacts 270×270 μm^2 in size. An In bump placed at the edge of the wafer was used as n-contact. The light emitted through the substrate was coupled into an integrating sphere for spectral and power analyses without any further measures taken to enhance light extraction. For structures grown on buffers with SiN_x interlayer, mesa-LEDs were fabricated and flip-chip mounted onto TO39 headers. Ni/Ag/Ni and V/Al/V/Au were used as p-contact and n-contacts, respectively. The size of the p-contact on the mesa-LED was 240×240 μm^2 .

3. DEFECT REDUCTION IN AlGaN BUFFER LAYERS

3.1 3D GaN nucleation

The in-situ reflectance measurement of a low-temperature GaN nucleation with the subsequent annealing and the overgrowth with AlGaN is shown in Figure 1. The different growth stages are indicated by the different shaded areas. Due to the higher refractive index of GaN compared to sapphire and the constructive interference which builds up between the optical wave reflected by the growing GaN surface and that by the GaN-sapphire interface, the reflectance increases during the deposition of the low-temperature GaN layer (stage I). As the temperature is raised during the annealing step carried out in stage II, the refractive index of GaN increases leading to a moderate increase in reflectance. However, as the temperature further increases, a sudden drop in reflectance can be observed due to the decomposition and roughening of the GaN surface resulting in the formation of three-dimensional nucleation islands. This is confirmed by the ex-situ AFM image of a similar sample taken after stage II (right side of Figure 1) showing GaN islands with a typical diameter and height of approximately 500 nm and 100 nm, respectively.

Within the first 150 nm of the Al_{0.2}Ga_{0.8}N buffer layer (stage III), the overgrowth is carried out at a reduced temperature of 1000°C to maintain a prolonged 3D growth mode. Consequently, the reflected intensity remains low. The AFM image taken after stage III illustrates the overgrowth and coalescence of the initial nuclei with a comparable large roughness.

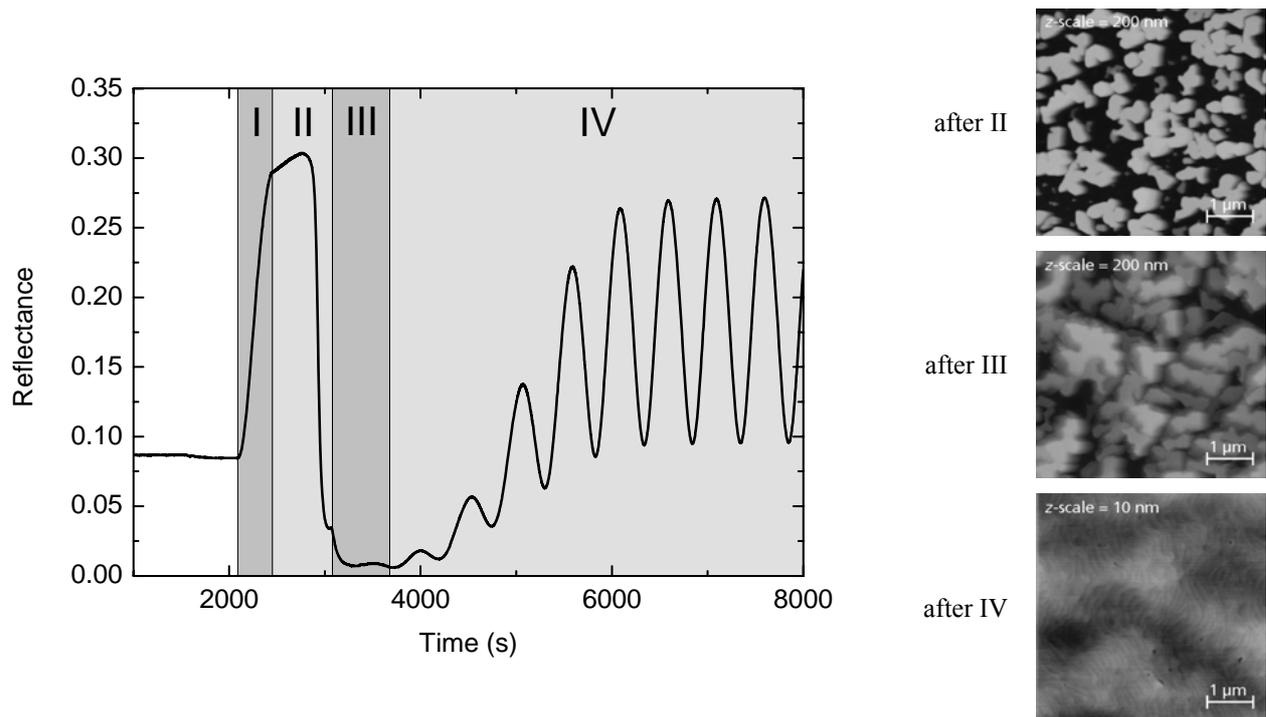


Figure 1. Left: In-situ reflectance signal during growth of AlGaIn on 3D GaN nucleation islands. The stages I-IV are indicated by the shaded areas and correspond to the deposition of the GaN nucleation layer, the annealing of the GaN nucleation layer, the initial growth of AlGaIn and the final growth of AlGaIn, respectively. Right: Corresponding ex-situ AFM images taken on samples after stages II, III, and IV, respectively.

However, as the growth temperature is raised to 1050°C in stage IV, the reflected light intensity increases continuously and pronounced interference oscillations appear which gain in amplitude over time. Obviously, the higher growth temperature leads to an enhanced lateral growth resulting in a fast coalescence of the islands and an effective flattening of the growing surface. After growth of roughly 1.5 μm, the surface is atomically flat as can be seen from the clearly resolved atomic steps in the AFM image taken after stage IV. The overall root mean square (RMS) roughness on a scale of 5×5 μm² is 0.4 nm.

An improved crystal quality of 1.5 μm thick AlGaIn buffer layers grown on the 3D GaN nucleation compared to the 2D AlGaIn nucleation can be inferred from a reduced full width at half maximum (FWHM) of the asymmetric 10.0 HRXRD reflection of 1000 arcsec for the 3D nucleation compared to 2340 arcsec for the 2D nucleation. Since the width of the asymmetric 10.0 HRXRD peak is mainly affected by lattice twist and is hence sensitive to edge-type threading dislocations [10], the 3D nucleation mode obviously leads to an effective reduction of the density of edge-type threading dislocations. The symmetric 00.2 peak width is reduced to a lesser extent by the 3D growth and only slightly improved from 610 to 470 arcsec. The FWHM values for the different AlGaIn buffer layers are summarized in Table 1.

3.2 SiN_x interlayer

An even more efficient approach for the reduction of the dislocation density in AlGaIn buffer layers is the deposition of SiN_x interlayers. The submonolayer coverage acts as a nano-masking where the overgrowth takes mainly place at the openings of the SiN_x interlayer. This selective growth leads to the formation of hexagonally shaped islands and, therefore, to a three-dimensional growth quite similar to the GaN nucleation. In addition to the defect reduction by the 3D growth mode, the nano-masks also block dislocations from propagating into the overgrown layer. As can be seen in the TEM image shown in Figure 2, most dislocations terminate at the SiN_x interlayer. Furthermore, in some areas the dislocations are bent and merged, creating bundles of dislocations and in between extended areas up to 1 μm in lateral dimension which are virtually free of dislocations.

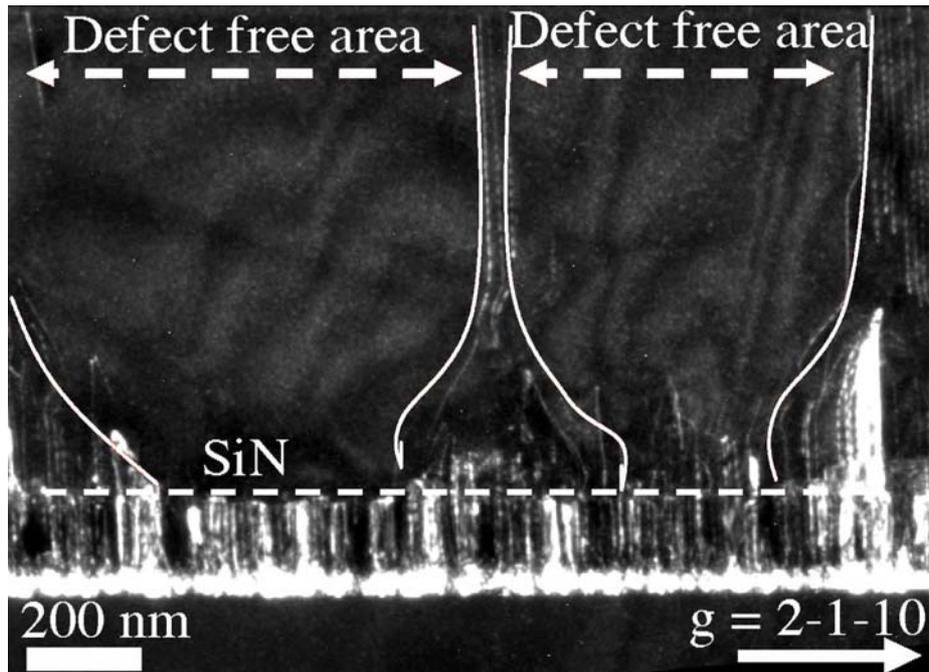


Figure 2. WBDF-TEM from cross section of an AlGaIn layer with SiN_x interlayer deposited 150 nm above the nucleation layer.

An improved dislocation reduction was observed when the SiN_x interlayer was deposited not directly on the nucleation layer, but after growth of 150 nm of AlGaIn. The optimal coverage was determined by the variation of the deposition time. Best results in terms of asymmetric HRXRD peak widths were achieved at a deposition time of 6 min. It was also observed that the surface roughening increased for higher deposition times and thus longer overgrowth times were required for the flattening of the surface. However, after growth of 2.5 μm AlGaIn, atomically flat surfaces were achieved in all cases. A detailed description of the growth parameter optimization can be found in [7].

Measuring the asymmetric 10.0 HRXRD reflections under the same conditions as on the 3D GaN and 2D AlGaIn nucleations, the peak width is determined to be as narrow as 510 arcsec. The symmetric 00.2 reflection peak width is reduced to 270 arcsec (see Table 1). These values underline the excellent crystal quality and the effective dislocation reduction achieved by the SiN_x nano-masks.

Table 1. FWHM values of the 00.2 and 10.0 HRXRD reflections for 1.5 μm thick AlGaIn buffer layers

	00.2	10.0
(1) 3D GaN nucleation	470 arcsec	1000 arcsec
(2) SiN _x interlayer	270 arcsec	510 arcsec
(3) 2D AlGaIn nucleation	610 arcsec	2340 arcsec

4. LED PERFORMANCE – ON-WAFER MEASUREMENTS

The integrated EL light output power as a function of the injection current (P-I characteristic) for 350-nm-LEDs grown on the three different buffer layers is plotted in Figure 3. These measurements have been carried out on-wafer collecting the light emitted through the substrate with no measures taken for improving light extraction or heat-sinking of the devices (the schematic of the measurement setup is depicted in the inset of Figure 3). The output power shows a continuous increase in the measured current range. However, plotting the external quantum efficiency (EQE) as a function of the injection current (not shown), a thermal rollover is noticeable already for currents exceeding 25 mA due to the self-heating of the devices. Hence, for a proper comparison, the light output power at an injection current of 20 mA was taken, which is 0.019 mW, 0.131 mW and 0.505 mW for the LEDs on the 2D nucleation, 3D nucleation and on the AlGaIn buffer with SiN_x interlayer, respectively. The corresponding EQEs are 0.027 %, 0.18 % and 0.71 %, respectively.

The increase in the optical output power by a factor of roughly 7 and 25 for the 3D GaN nucleation and the SiN_x interlayer compared to the 2D AlGaIn nucleation is mainly attributed to an enhanced internal quantum efficiency due to the reduction of the edge-type dislocation density by these two approaches. This is supported by excitation-density and temperature dependent PL measurements showing a clear enhancement of the internal quantum efficiency for LED structures grown on these defect-reduced AlGaIn buffers [9].

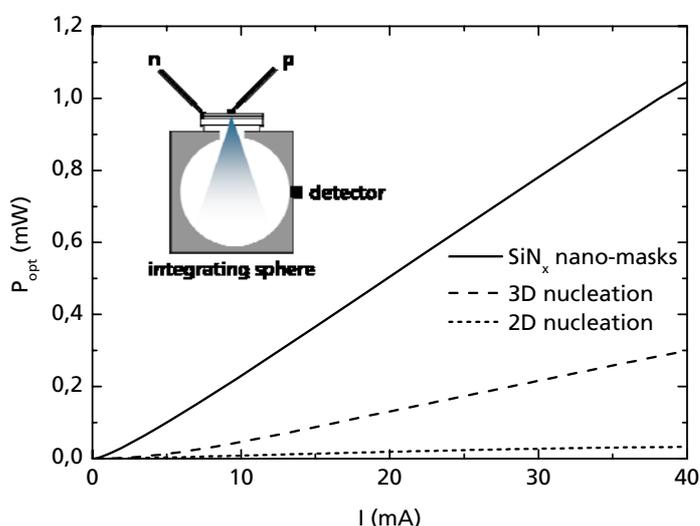


Figure 3. Optical output power of LED structures grown on SiN_x interlayer (solid), 3D (long-dashed) and 2D nucleation layer (short-dashed) as a function of the injection current. Measurements were taken on-wafer without any measures to improve light extraction and heat dissipation. The schematic of the measurement setup is depicted in the inset.

5. LED PERFORMANCE – FLIP-CHIP MOUNTED MESA-DEVICES

Based on the results of the on-wafer EL measurements, mesa-LEDs were fabricated from layer sequences grown on AlGaIn buffer structures with SiN_x interlayer. The LED chips were flip-chip mounted onto TO39 headers for improved heat dissipation. As can be seen in Figure 4, excellent current-voltage characteristics were achieved with a reverse bias current below 10 nA at -5 V. The forward voltage at a current of 20 mA is as low as 3.8 V, underlining the high conductivity of the n- and p-AlGaIn layers as well as the low resistance of the applied metal contacts [11].

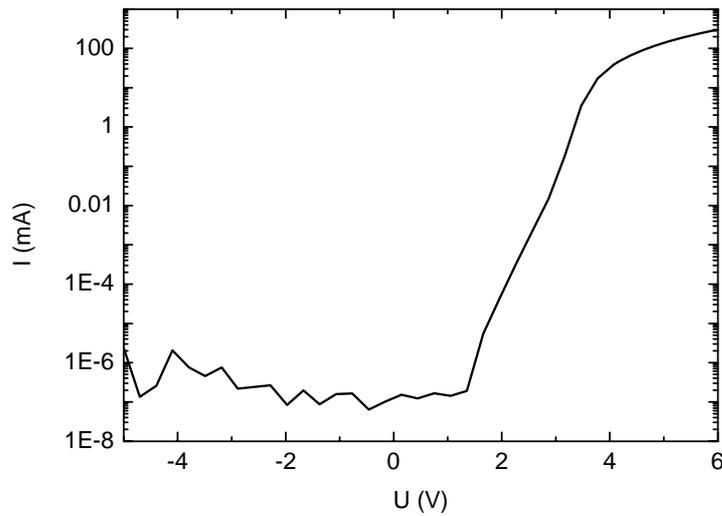


Figure 4. Current-voltage characteristics on a logarithmic scale of a 350-nm-emitting mesa-LED grown on SiN_x interlayer. The reverse current is below 10 nA at -5 V. The forward current at 20 mA is 3.8 V and rises to 6 V at 300 mA.

The emission spectrum of the LED taken at an injection current of 20 mA is shown in Figure 6. The peak wavelength corresponding to the QW emission is located at 348 nm with a FWHM of 7.5 nm. It is clearly visible that the defect luminescence in the yellow-green spectral range is at least 3 orders of magnitude lower.

Figure 5 presents the light output power as a function of the injection current. Due to the improved heat dissipation compared to the on-wafer measurements, the light output power linearly increases up to 100 mA before a noticeable downward bending of the P-I characteristic due to device heating can be observed. At this current, the light output power is 5.5 mW, corresponding to a maximum EQE of 1.55 %. The output power reaches its maximum of 12.2 mW at a current of 300 mA, limited by thermal rollover.

The improvement in the maximum EQE of the fully packaged mesa-LED is mainly attributed to an increased light extraction by the additional side facets. A comparison of the output power achieved on-wafer and of the flip-chip mounted mesa-LEDs within the linear regime (where the self-heating of the device can be neglected) indicates an improvement in light extraction of a factor of 2. Besides, the improved heat dissipation enables the operation at higher current densities. Consequently, the higher carrier density in the QW region leads to a saturation of the defect-induced non-radiative recombination and, hence, to a further increase in the maximum EQE.

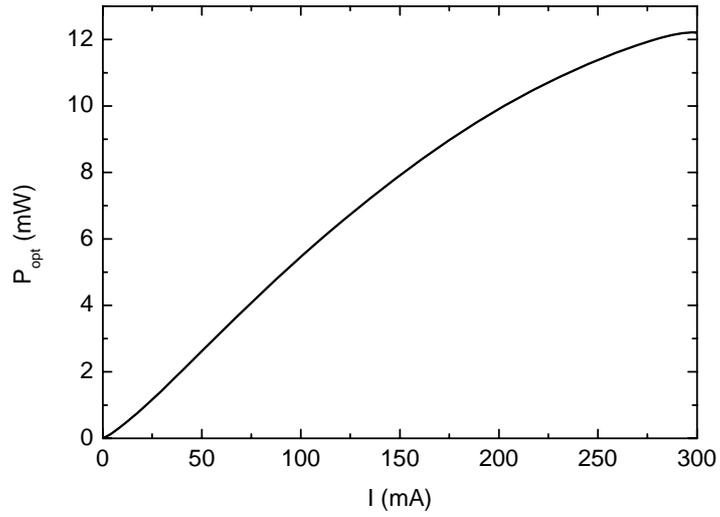


Figure 5. Optical output power of a mesa-LED emitting at 348 nm grown on SiN_x interlayer as a function of the injection current. At currents exceeding 100 mA, a thermal rollover becomes observable.

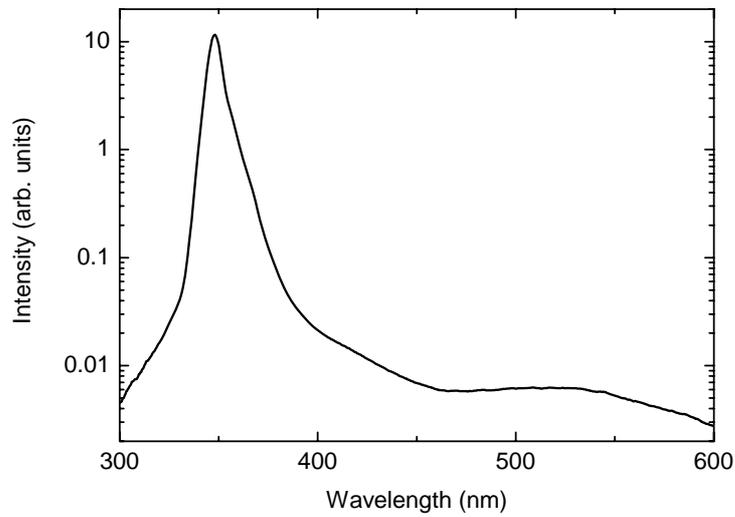


Figure 6. EL spectrum on a logarithmic intensity scale of a mesa-LED grown on SiN_x interlayer. The peak wavelength is located at 348 nm. Defect luminescence in the yellow-green range is more than 3 orders of magnitude less intense than the UV emission.

6. SUMMARY

We reported on two different approaches for the defect reduction in $\text{Al}_{0.2}\text{Ga}_{0.8}\text{N}$ buffer layers for the realization of 350-nm-emitting UV-LEDs with improved efficiency. Both the application of a 3D GaN nucleation and of a SiN_x interlayer lead to narrower HRXRD ω -scan peak widths indicative of decreased threading edge-type dislocation densities. Atomically flat surfaces were achieved enabling these layers to serve as buffers for 350-nm-LEDs. The beneficial effect of the reduced dislocation density on the optical output power of these LEDs is demonstrated by on-wafer measurements. Best results were achieved on LEDs grown on SiN_x interlayers which showed an increase in light output power by a factor of 25 compared to structures without special defect reduction. On these buffer layers, flip-chip mounted mesa-LEDs have been realized with a low forward voltage of 3.8 V at 20 mA and a maximum output power of 12.2 mW at 300 mA.

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REFERENCES

- [1] Khan, A., Balakrishnan, K. and Katona, T., “Ultraviolet light-emitting diodes based on group three nitrides”, *Nat. Photonics* 2(2), 77-84 (2008).
- [2] Chen, K. X., Dai, Q., Lee, W., Kim, J. K., Schubert, E. F., Grandusky, J., Mendrick, M., Li, X. and Smart, J. A., “Effect of dislocations on electrical and optical properties of n-type $\text{Al}_{0.34}\text{Ga}_{0.66}\text{N}$ ”, *Appl. Phys. Lett.* 93(19), 192108 (2008).
- [3] Tsuzuki, H., Mori, F., Takeda, K., Ichikawa, T., Iwaya, M., Kamiyama, S., Amano, H., Akasaki, I., Yoshida, H., Kuwabara, M., Yamashita, Y. and Kan, H., “High-performance UV emitter grown on high-crystalline-quality AlGaIn underlying layer”, *Phys. Status Solidi A* 206(6), 1199 (2009).
- [4] Schenk, H. P. D., Vennegues, P., Tottereau, O., Riemann, T. and Christen, J., “Three-dimensionally nucleated growth of gallium nitride by low-pressure metalorganic vapour phase epitaxy”, *J. Cryst. Growth* 258(3-4), 232 (2003).
- [5] Hertkorn, J., Lipski, F., Brückner, P., Wunderer, T., Thapa, S. B., Scholz, F., Chuvilin, A., Kaiser, U., Beer, M. and Zweck J., “Process optimization for the effective reduction of threading dislocations in MOVPE grown GaN using in situ deposited SiN_x masks”, *J. Cryst. Growth* 310(23), 4867 (2008).
- [6] Gutt, R., Kirste, L., Passow, T., Kunzer, M., Köhler, K. and Wagner, J., “Reduction of the threading edge dislocation density in AlGaIn epilayers by GaN nucleation for efficient 350 nm light emitting diodes”, *Phys. Status Solidi B* 247(7), 1710 (2010).
- [7] Forghani, K., Klein, M., Lipski, F., Schwaiger, S., Hertkorn, J., Leute, R. A. R., Scholz, F., Feneberg, M., Neuschl, B., Thonke, K., Klein, O., Kaiser, U., Gutt, R. and Passow, T., “High quality AlGaIn epilayers grown on sapphire using SiN_x interlayers”, *J. Crystal Growth*, in press, [doi:10.1016/j.jcrysgro.2010.08.044](https://doi.org/10.1016/j.jcrysgro.2010.08.044)
- [8] Klein, O., Biskupek, J., Kaiser, U., Forghani, K., Thapa, S. B. and Scholz, F., “Simulation supported analysis of the effect of SiN_x interlayers in AlGaIn on the dislocation density reduction”, *Journal of Physics: Conference Series* 209(1), 012018 (2010).
- [9] Kunzer, M., Gutt, R., Kirste, L., Passow, T., Forghani, K., Scholz, F., Köhler, K. and Wagner, J., “Improved quantum efficiency of 350 nm LEDs grown on low dislocation density AlGaIn buffer layers”, submitted to IWN Proceedings 2010 and *Physica Status Solidi*, in press
- [10] Moram, M. A. and Vickers, M. E., “X-ray diffraction of III-nitrides”, *Reports on Progress in Physics* 72(3), 036502 (2009).
- [11] Passow, T., Gutt, R., Maier, M., Pletschen, W., Kunzer, M., Schmidt, R., Wiegert, J., Luick, D., Liu, S., Köhler, K. and Wagner, J., “Ni/Ag as low resistive ohmic contact to p-type AlGaIn for UV LEDs”, *Proc. SPIE Vol.* 7617, 76171I (2010).