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LEDs on HVPE grown GaN substrates: Influence of macroscopic surface features

SK. S. Rahman,[1] R. A. R. Leute,[1] J. Wang,[1] T. Meisch,[1] M. Klein,[1] F. Scholz,[1] K. Koyama,[2] M. Ishii,[2] and H. Takeda[2]
[1]*Institut für Optoelektronik, Universität Ulm, 89069 Ulm, Germany*
[2]*Namiki Precision Jewel Co., Ltd., 8-22, Shinden 3-chome, Adachi-ku, Tokyo 123-8511, Japan*

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We demonstrate the strong influence of GaN substrate surface morphology on optical properties and performance of light emitting devices grown on freestanding GaN. As-grown freestanding HVPE GaN substrates show excellent AFM RMS and XRD FWHM values over the whole area, but distinctive features were observed on the surface, such as macro-pits, hillocks and facets extending over several millimeters. Electroluminescence measurements reveal a strong correlation of the performance and peak emission wavelength of LEDs with each of these observed surface features. This results in multiple peaks and non-uniform optical output power for LEDs on as-grown freestanding GaN substrates. Removal of these surface features by chemical mechanical polishing results in highly uniform peak wavelength and improved output power over the whole wafer area. [http://dx.doi.org/10.1063/1.4890348]

I. INTRODUCTION

Gallium nitride (GaN) is an extremely promising material for optoelectronic devices such as light-emitting diodes (LEDs) and laser diodes (LDs).[1,2] GaN based white LEDs for solid state lighting are increasingly deployed due to their low energy consumption and very long lifetime.[3] State-of-the-art GaN based devices are typically grown heteroepitaxially on foreign substrates with metalorganic vapour phase epitaxy (MOVPE). GaN layers on such foreign substrates, typically sapphire, suffer from a high dislocation density ($10^8 - 10^{10}$ cm^{-2}) which leads to many drawbacks such as the increase of the leakage currents in blue and green LEDs, reduced efficiency of near-UV LEDs and decreased lifetime of laser diodes.[4,5] Therefore, homo-epitaxy is preferable. With GaN bulk crystal growth still in its infancy, most of the bulk substrates are based on thick GaN layers grown by hydride vapor phase epitaxy (HVPE) separated from foreign substrates.[6–8] These free-standing GaN layers grown by HVPE offer an excellent crystal quality and low threading dislocation densities[9,10] with low AFM roughness. However, the fast HVPE growth of thick layers, even with homogeneously high crystal quality, on MOVPE templates introduces defects and pits which lead to a macroscopically rough surface. This interface between the pseudo bulk substrate and the subsequent MOVPE grown layers may be therefore critical for the device performance. In this work, we investigated the performance of test structures grown on freestanding as-grown substrates compared to polished substrates in order to identify the effects of macroscopic surface features and how the excellent crystalline quality of the GaN substrates can be used for high performance devices.

II. EXPERIMENTAL

The growth of freestanding GaN substrates was performed in a commercial Aixtron single-wafer HVPE system with a horizontal quartz-tube, heated in a furnance with five zones. The detailed growth conditions for the 1 mm thick self-separated freestanding GaN substrates can be found elsewhere.[11] The investigated test LED structures were grown on as-grown freestanding GaN substrates and also

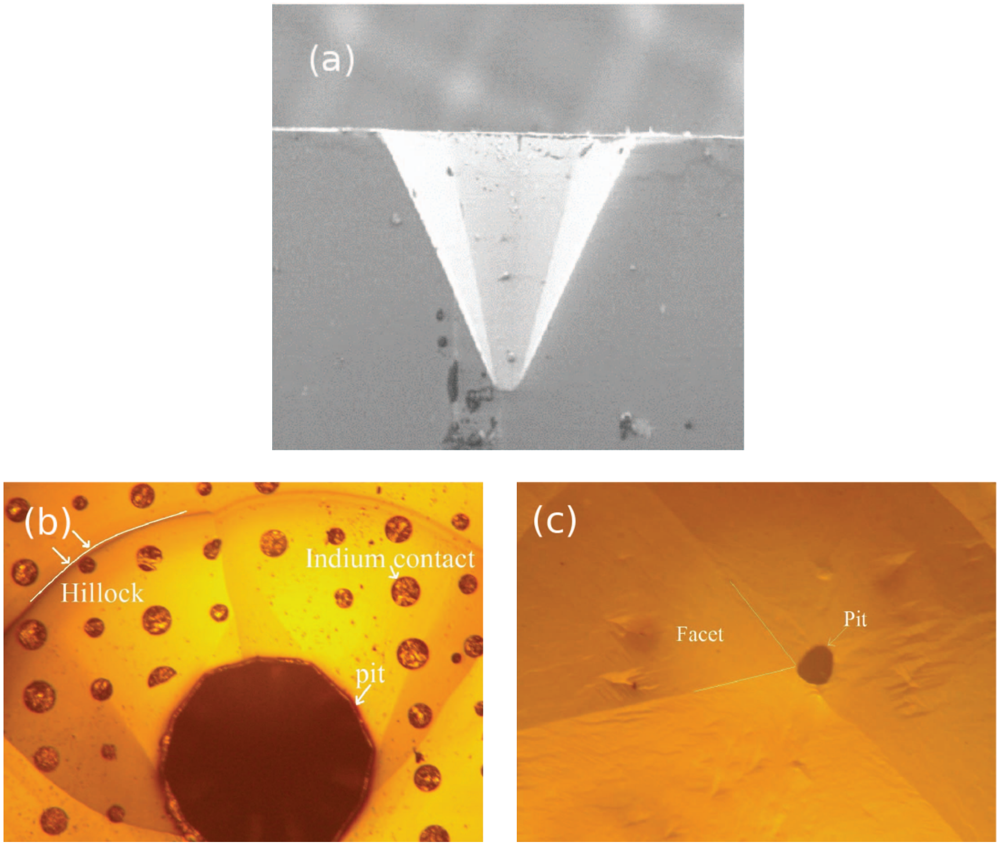

FIG. 1. Surface features of LEDs grown on freestanding GaN, (a): Cross-sectional SEM image of a macro-pit, (b): Nomarski microscopic image of hillocks with evaporated contacts. (c): Nomarski microscopic image of facets.

on polished freestanding GaN substrates using an Aixtron 200/4 RF-S metal organic vapour phase epitaxy (MOVPE) system. The LED structures consisted of a 1 μm undoped GaN buffer layer with a 1.5 μm Si doped n-type GaN layer on top. Then a 50 nm thick InGaN pre-well layer was grown. The active region consisted of five 2.6 nm thick $In_{0.09}Ga_{0.91}N$ QWs separated by 12 nm thick GaN barriers and afterwards, a 5 nm GaN capping layer was deposited. A 15 nm thick $Al_{0.1}Ga_{0.9}N$ electron blocking layer (EBL) was grown on top of the active region followed by a 60 nm Mg doped p-type GaN layer. The samples were characterized by scanning electron microscopy (SEM), atomic force microscopy (AFM), Nomarski microscopy and electroluminescence (EL) measurements. In order to perform the EL measurements, p and n contacts were realized by lithographic steps with indium deposition. All EL measurements were carried out on-wafer at room temperature with a fixed p-contact size of 140 μm diameter. We measured p-contacts over the whole area of the samples to probe different positions on surface. The intensity of the LED light is collected inside an integrating sphere by a photo diode and a spectrometer is used to record the spectra.

III. RESULTS

The surfaces of the as-grown HVPE freestanding GaN and the subsequently grown structures such as QWs and LEDs show macroscopically uneven surfaces. Three distinct macroscopic features were revealed on both the surfaces of HVPE freestanding GaN layer and the LED test structures grown thereon: i) macro size V-shaped pits (a cross-section of a pit is shown in figure 1(a), which has a diameter of around 700 μm and the height is about 550 μm). Macro pits of different sizes (typically 100–1000 μm diameter) were observed on top of the surface with a density in the mid 10^1 cm^{-2}. ii) surrounding the pits, hillocks were observed which have an average diameter of nearly

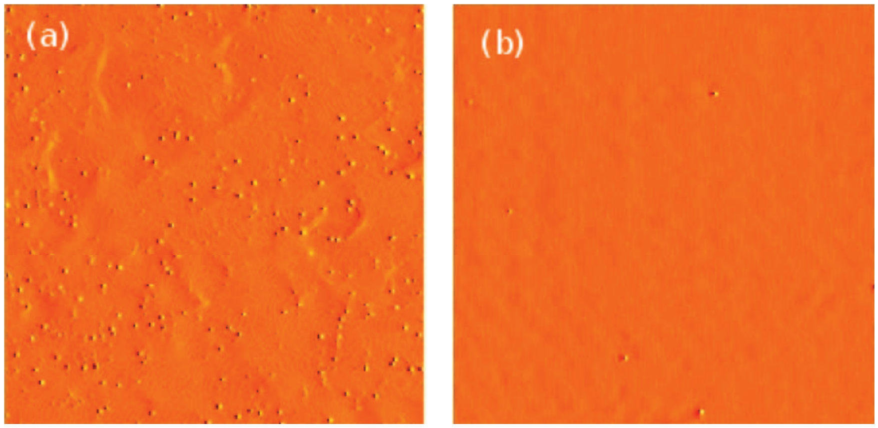

FIG. 2. AFM micrographs of $10 \times 10\ \mu m^2$ scan size of QWs grown on different substrates. InGaN overgrowth of dislocations leads to a decoration with V-pits. AFM analysis yields average threading dislocation densities of 10^8 cm^{-2} (a) and 5×10^6 cm^{-2} (b) for QWs grown on sapphire and freestanding GaN substrates, respectively.

2-3 mm (Fig. 1(b)). iii) hexagonally symmetric facets were also found (Fig. 1(c)). Typically these facets start laterally from a pit and extend over several millimeters. It should be noted, that all of these macroscopic surface features developed on the HVPE freestanding GaN sates during the growth in the HVPE system and these surface modifications originate from pits. The origin of formation of such pits in the HVPE freestanding GaN substrates is not clear yet. However, the HVPE GaN layers as well as the subsequently grown structures were very smooth on a microscopic scale. An atomically flat surface was revealed on LEDs grown on the as-grown freestanding GaN substrates and the RMS roughness is 0.23 nm in a $10 \times 10\ \mu m^2$ scan size measured on a hillock site. Besides, very low threading dislocation densities (mid 10^6 cm^{-2}) were found on the test structures grown on the as-grown freestanding GaN substrates (Fig. 2). So we consider the crystalline quality to be very high, even on the hillocks and facets. LED contacts have been processed on these features in order to perform EL spectrum measurements on these surface features. The measurements were carried out at room temperature. Fig. 3(a) shows the EL spectra of a LED at different injection currents for a contact on a relatively flat area, where the intended peak emission at 432 nm appeared as dominant peak. We observe the highest output power (3.6 mW at 20 mA) for contacts in this region. At low injection currents, a peak at 490 nm was present and at higher injection currents a 3rd peak at 462 nm was also observed. This behavior is observed on approximately 25% of the sample surface. Fig. 3(b) shows the EL spectra of the same sample measured with the p-contact on a facet as shown in Fig. 1(c). The intended peak emission around 432 nm was completely missing, but interestingly, the EL spectra at low injection currents on the facet showed 462 nm peak wavelength. With increasing injection currents a second peak appeared nearly at 490 nm. At even higher injection currents the intended peak emission at 432 nm was observed as a little shoulder. However, on this facet the emission at 462 nm remained dominant. We also observed that this 462 nm peak emission was fairly constant throughout the facet. On the other hand, the dominant peak emission on a hillock was observed at 411 nm (Fig. 3(c)). Moreover, here, a 2nd peak appeared at 462 nm with a little shoulder at 432 nm. The EL spectra indicate indium non-uniformity in the QWs. It seems that on the facets, indium incorporation is favorable, whereas on a hillock, indium incorporation is more unfavorable and the intended peak emission was only found on the flat areas. Photoluminescence measurements (not shown) confirm these findings. These results clearly demonstrate that the surface has a significant effect on the emission properties of LEDs and also has a great influence on the indium incorporation during the growth of the QWs ([12]). In order to separate the effects of the surface topology from the crystal quality of the underlying material, an optimized chemical mechanical polishing (CMP) treatment was applied on some freestanding GaN samples. We achieve a macroscopically as well as atomically smooth surface, when the Ga-polar surface was polished with a silica based slurry after mechanical polishing.[13] During mechanical

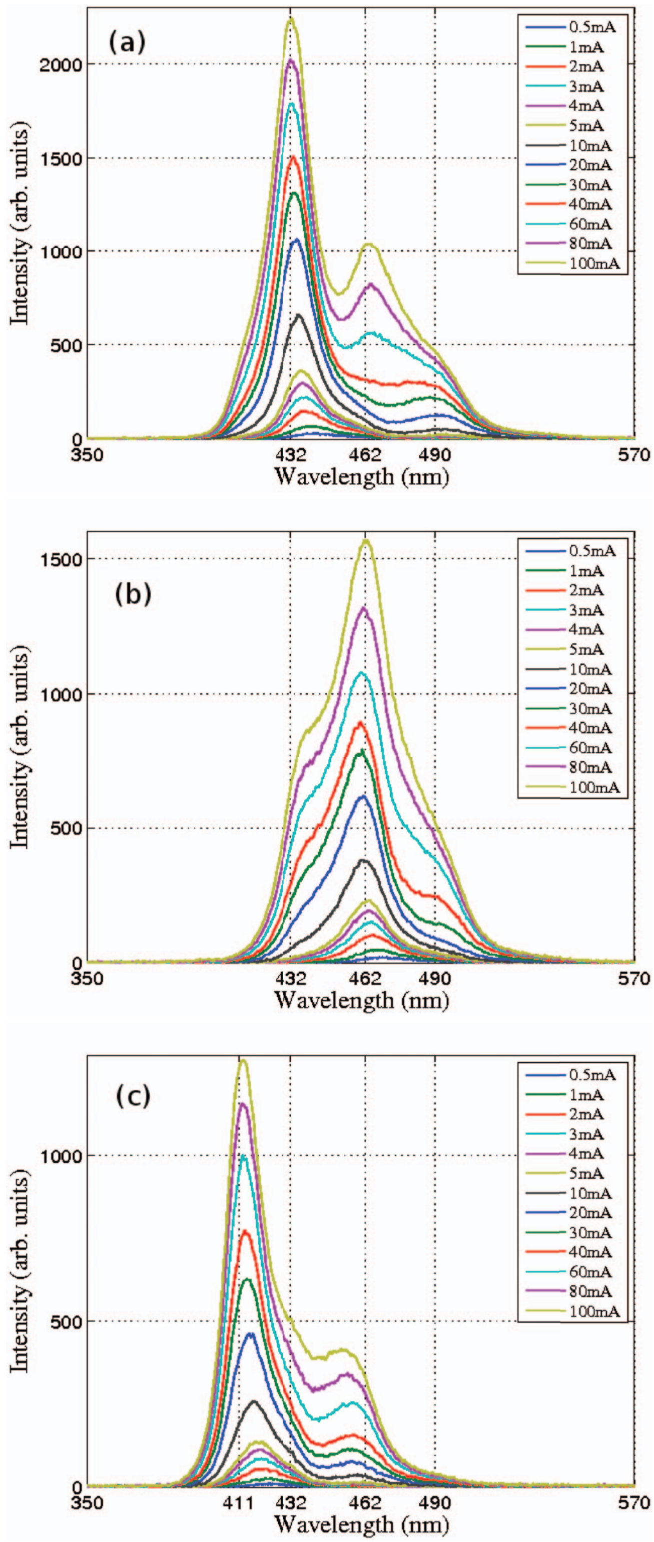

FIG. 3. EL spectra at different driving currents of LEDs grown on as-grown freestanding GaN substrate. (a) represents the EL spectra on a relatively flat area, (b) represents the EL spectra on a facet and (c) depicts the EL spectra on a hillock.

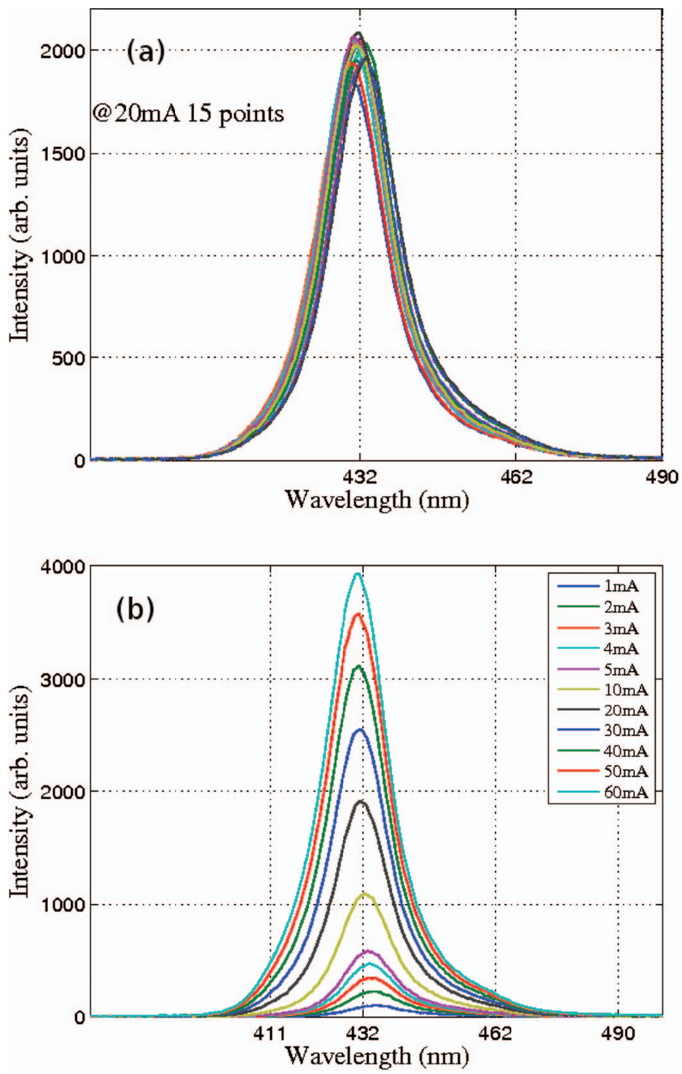

FIG. 4. EL spectra of LEDs grown on polished freestanding GaN substrate. (a) 15 EL spectra at 20 mA current and measured at 15 different areas of the sample and (b) EL spectra at different driving currents

polishing, ≈250 μm were removed to create an even surface. Afterwards, the surface features such as hillocks and facets were removed, but pits with reduced diameter are left, as their depth exceeds the thickness of the removed layer. This allows us to probe areas around pits which exhibited surface modifications in the as-grown state. The root mean square (RMS) of the surface roughness was found to be around 0.9 nm. Typically the mechanical polishing can create sub-surface damage. However, after CMP, no damage is visible in cathodoluminescence (CL) imaging.[13] Afterwards, similar LED structures were grown on these polished freestanding GaN substrates. Fig. 4(a) represents the EL spectra at 20 mA driving current of LEDs grown on such polished freestanding GaN substrates. The measurements were taken at 15 different areas of the sample. All spectra showed a single peak at 430 nm with neither wavelength fluctuations nor multiple peaks observable. Furthermore, equally stable behavior is found for different injection currents for these LEDs grown on polished freestanding GaN substrates (fig. 4(b)). This homogeneous peak emission was found throughout the sample. Moreover, they exhibit very homogeneous and improved optical output power (Fig. 5(a)). For comparison, Fig. 5(b) represents the L-I characteristics of LEDs grown on as-grown freestanding GaN substrates. Here we observed non-uniform optical output power over the entire surface areas of the sample. We also found that the polishing caused a noticeable improvement of the optical

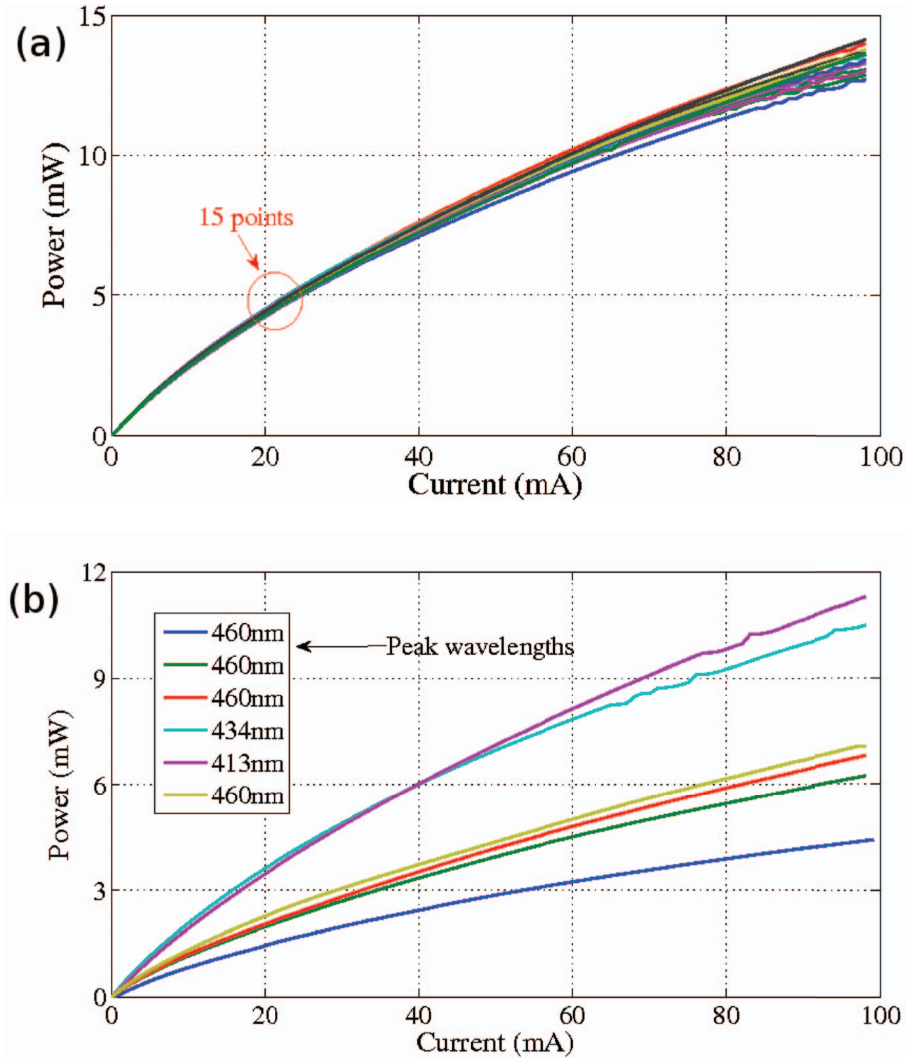

FIG. 5. L-I characteristics of LEDs grown on (a) polished freestanding GaN substrates where 15 points showed homogeneous output power and identical peak wavelengths and (b) on as-grown freestanding GaN substrates where non-uniform output power and peak emission was observed.

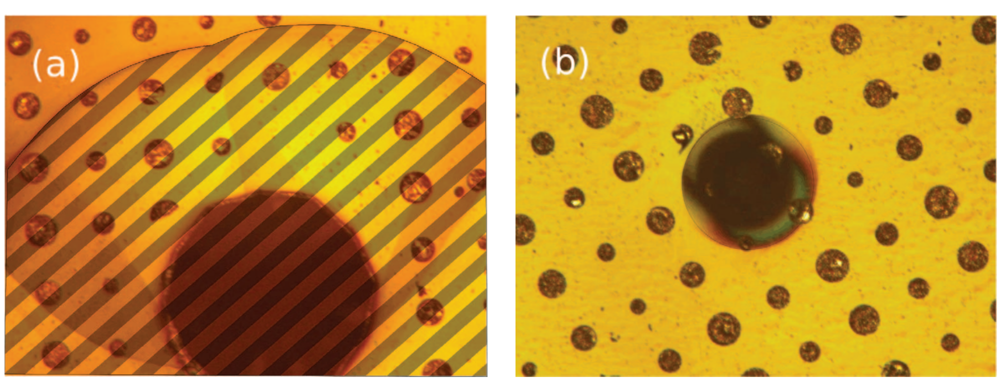

FIG. 6. Nomarski image of area surrounding pit. On the as-grown substrate (a), low power and multiple emission peaks are found in the marked area. On polished substrate (b), all contacts, right to the very vicinity of the pits show high output power and uniform emission wavelength.

output power of LEDs. Before applying the surface treatment on the HVPE GaN surface, the highest optical output power of LEDs was found to be about 3.6 mW at 20 mA driving current on flat areas of the sample which comprised roughly 25 %. After polishing, over 4.6 mW optical output power at 20 mA injection current was measured uniformly over the full area of the sample. There is no deviation around the pits, where, in contrast, the as-grown samples are strongly influenced (see fig. 6). Only contacts evaporated directly onto a pit (less than 1 % of the area) did not perform this well. This clearly corroborates our finding that the reason of peak wavelength fluctuations of the devices grown on as-grown freestanding GaN substrates was due to the presence of macroscopic defects such as hillocks and facets on the as-grown HVPE freestanding GaN surface. These surface features significantly alter and impair the device performance despite excellent XRD FWHM and AFM RMS values. Especially, we found out that even on large flat areas outside the pit related features, the EL exhibit the intended peak emission, still showed multiple peak behavior and also the optical output power was not homogeneous throughout the sample. However, these effects are solely connected to the as-grown surface and not to the underlying material. They can be remedied on nearly 100 % of the area by applying polishing, yielding uniform emission and high output power.

IV. CONCLUSION

In conclusion, we demonstrated the effects of macroscopic surface defects on the performances of opto-electronic devices. Three distinctive surface features were revealed on the surface of HVPE freestanding GaN surface such as macro-pits, hillocks and facets. EL measurements showed that these hillocks and facets have a great impact on the emission properties of LEDs. We observed that on the facets, the peak emission wavelength was increased by nearly 30 nm and remains fairly uniform throughout the facet. This increased peak wavelength might indicate that the indium incorporation is favorable on such a facet. We also observed that on the hillocks the indium incorporation appears unfavorable. The optical output power was non-uniform over the entire surface of the LEDs grown on the as-grown freestanding GaN substrates, even in far distance of these surface features. Using an optimized surface treatment, i.e. CMP, the macroscopic surface features were removed. The LEDs grown on the polished freestanding GaN substrates showed excellent homogeneity in terms of the optical output power and the peak emission wavelength. With the application of CMP on the surface of HVPE freestanding GaN, nearly 100 % of the sample surface yields high performance LEDs, indicating that the observed features are solely a disturbation of the surface and not an extension of crystal defects within the layer. Therefore, the high crystalline quality of the HVPE grown GaN substrates can only be utilized to its fullest when the surface is carefully and appropriately treated.

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