MOVPE Growth of Semipolar GaN on Patterned Sapphire Wafers: Growth Optimisation and InGaN Quantum Wells

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We are able to achieve two different GaN semipolar surfaces by growing on patterned sapphire substrates: $(10\bar{1}1)$ *GaN on* $(11\bar{2}3)$ *sapphire and* $(11\bar{2}2)$ *on* $(10\bar{1}2)$*. By this approach, the growth of a coalesced semipolar layer on a large area of about two inches diameter is possible. Well known from the c-plane direction, an in-situ deposited SiN interlayer could reduce the defect density also in semipolar directions. The influence of position and growth temperature of the SiN layer on the GaN layer quality was investigated. Using optimized growth conditions, the full width at half maximum (FWHM) of X-ray rocking curve peaks could be halved. Moreover, an optimized temperature profile during growth of the main layer helps to improve the crystal quality even further. First InGaN quantum wells (QWs) were deposited on these optimized semipolar surfaces. When transferring the c-plane QW growth conditions to* $(11\bar{2}2)$ *GaN on* $(10\bar{1}2)$ *sapphire, a lower growth rate and a reduced indium incorporation was observed. By using thicker quantum wells grown with a higher indium flux, the emission wavelength could be shifted to 500 nm. In the case of* $(10\bar{1}1)$ *GaN on* $(11\bar{2}3)$ *sapphire, high-quality quantum wells with an emission wavelength of about 525 nm have been achieved.*

1. Introduction

Most of the common light emitting diodes or laser diodes based on InGaN/GaN are grown in the well-known c (0001) direction. On the one hand, excellent crystal quality can be obtained in such a direction, but on the other hand, the crystal symmetry of GaN causes strong piezoelectric fields within such heterostructures. These internal electric fields bend the energy levels leading to a charge carrier separation in the quantum wells. The reduced overlap of the wavefunctions of electrons and holes leads to a reduced recombination probability — the efficiency of a light emitting device decreases. In addition, a red-shifted emission wavelength as a result of a reduced effective bandgap is observable, known as quantum confined Stark effect (QCSE). To avoid or to reduce these internal electric fields, the growth in non-c-directions seems to be a promising way. However, growth in a non-polar direction is typically affected by a low crystal quality [1, 2]. Semipolar growth directions are a good compromise between an acceptable crystal quality and reduced QCSE.

One very interesting technique to grow GaN in the well-known c-direction obtaining yet a semipolar surface was presented for example by Okada et al. [3]: They patterned r-plane sapphire substrates by etching trenches with c-like side facets into the wafer. With the help of selective epitaxy, crystal growth just on the c-facets is possible. At the beginning, GaN forms triangular shapes which eventually coalesce to a planar semipolar surface (cf. Fig. 1) after a suitable growth time.

Our investigations are focused on two different semipolar directions: $(10\bar{1}1)$ GaN on $(11\bar{2}3)$ patterned sapphire substrates and $(11\bar{2}2)$ GaN on $(10\bar{1}2)$ sapphire.

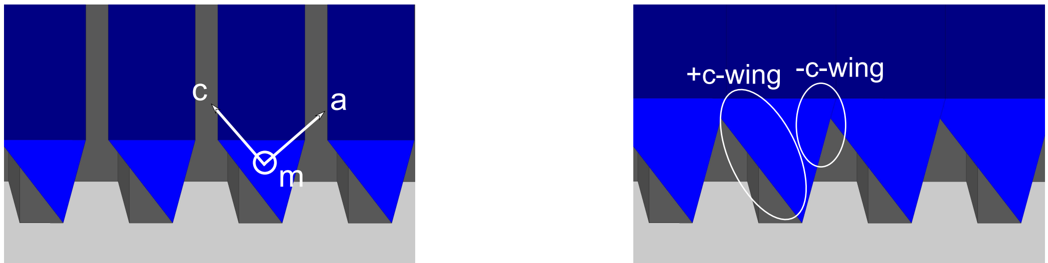


Fig. 1: Growth of GaN stripes (blue) with triangular shapes on a patterned substrate (grey). The triangular stripes (left) eventually coalesce to a planar surface (right).

2. Experimental

In order to pattern the sapphire substrate with trenches having a c-plane-like side facet, at first, a 200 nm thick layer of SiO_2 is deposited, which later acts as a mask for selective area growth. It is followed by an about 500 nm thick nickel layer structured via optical lithography with a stripe mask (3 µm opening, 6 µm period). The pattern is transferred into the sapphire by reactive ion etching. The resulting grooves have a width at the bottom of about 1.5 µm, a depth of about 1.2 µm and possess a c-plane-like facet.

All samples investigated in this study were grown in a low-pressure horizontal metal organic chemical vapor phase epitaxy (MOVPE) reactor with the precursors trimethylgallium (TMGa), trimethylaluminium (TMAl) and high purity ammonia. The growth starts with an oxygen-doped AlN nucleation layer at low temperature (about 960 °C), followed by a GaN buffer layer at about 1070 °C and a thick GaN top layer at 1030 °C. The reactor pressure was set to 150 mbar and the V/III ratio was 650. The crystal quality of the GaN layer was investigated by high resolution X-ray diffraction (HRXRD) and photoluminescence (PL) measurements. The surface morphology was mainly evaluated by scanning electron microscopy (SEM).

2.1 Growth optimisation

As already known from the growth of common c-plane GaN, an *in-situ* deposited thin SiN interlayer could reduce the defect density considerably (cf. Hertkorn et al. [4]). Similar as in an epitaxial lateral overgrowth procedure with an *ex-situ* deposited SiO_2 and lithographically structured mask, a great deal of the vertically running dislocations are stopped by the mask, and the remaining defects my be bent in a lateral direction. In the case of our semipolar oriented GaN, the SiN interlayer is deposited after a thin high-temperature GaN buffer layer grown at about 1080 °C. Then, about 4 μm GaN are deposited at 1000 °C leading to full coalescence of the stripes. The corresponding photoluminescence spectra (cf. Fig. 2) for $(11\bar{2}2)$ samples show a significant improvement of the crystal quality. By inserting the SiN nanomask layer, a doubling of the D^0X peak intensity at about 3.48 eV was achieved (cf. Fig. 2, left part), while the intensity of the stacking fault related signals at an energy of 3.42 eV decreased significantly (cf. Fig. 2, right part).

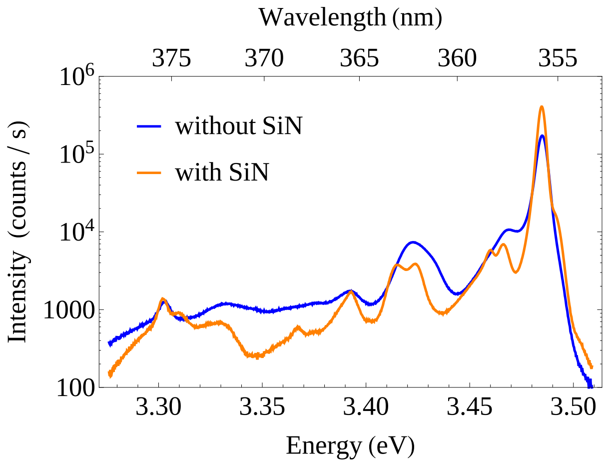


Fig. 2: Photoluminescence spectra of $(11\bar{2}2)$ GaN with (orange) and without (blue) SiN interlayer. Introducing a SiN interlayer improves the crystal quality significantly, visible in a much higher D^0X intensity and a reduced intensity of the defect emission band (for example at 3.42 eV).

X-ray measurements of the symmetric $(11\bar{2}2)$ and asymmetric $(11\bar{2}4)$ reflections confirm the improvement of crystal quality (cf. Fig. 3). In particular, the FWHM of the asymmetric reflection decreases significantly.

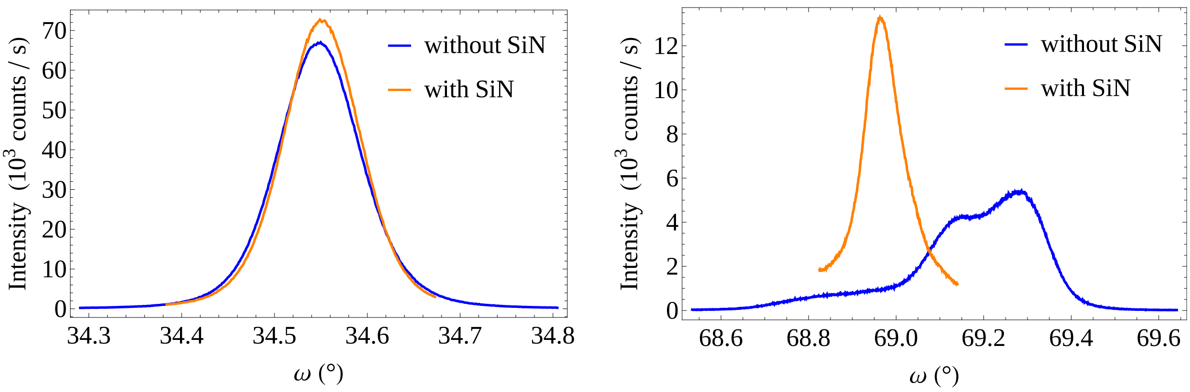


Fig. 3: Rocking curve (RC) measurements of $(11\bar{2}2)$ GaN in $(11\bar{2}2)$ (left) an $(11\bar{2}4)$ (right) direction. The intensity of symmetric reflections (right) increases just slightly by inserting a SiN interlayer. However, asymmetric reflections (right) show a significant crystal quality improvement due to the SiN interlayer.

The influence of such a SiN interlayer on the growth of $(10\bar{1}1)$ GaN was also investigated.

Using comparable growth conditions as for $(11\bar{2}2)$, the FWHM of the symmetric $(10\bar{1}1)$ X-ray reflection decreases from 770 arcsec to 510 arcsec. Although we have carefully studied further modifications of the growth conditions — like depositing temperature, thickness of buffer layer, etc. — we could not obtain a clear additional improvement. Previous cathodoluminescence (CL) investigations of Schwaiger et al. [5] showed that the −c-wing of our stripes (cf. Fig. 1) exhibits a higher defect density than the +c-wing. To take advantage of this given fact, an accelerated coalescence of the individual stripes could help to achieve a further crystal quality improvement. As shown in Fig. 4, for $(11\bar{2}2)$ layers, the +c-wing grows much faster than the −c-wing at first. By adding a cold top layer, the +c-wing coalesces with the left part of the adjacent +c-wing, whereas the growth of the −c-wing stops in a “dead end”.

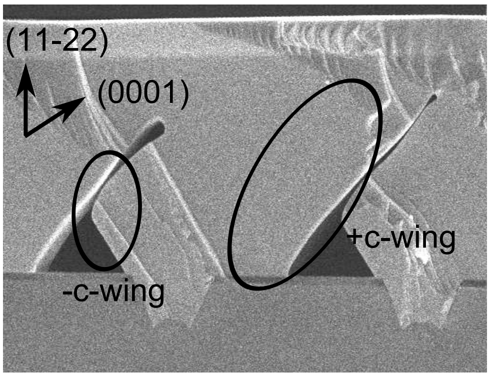


Fig. 4: Cross section SEM image of $(11\bar{2}2)$ GaN. Using slightly higher temperatures at the beginning of the growth, the stripes are not able to coalesce; a clear +c-wing and −c-wing, respectively, is visible. By adding a top layer, grown at reduced temperatures, the +c-wing gets pushed and overgrows the −c-wing, which exhibits an higher defect density. Due to the cold GaN top layer, the total defect density at the semipolar surface could be reduced significantly.

This effect may be pushed by growing a top layer at slightly reduced temperature. Therefore, we have grown three samples with different top layer temperatures. For temperatures of 1000 °C and 940 °C, we observe a reduced and broadened D^0X signal in PL (cf. Fig. 5), whereas a clear optimum is visible at a temperature of 970 °C. By this technique, the FWHM of the $(11\bar{2}2)$ rocking curves could be reduced to less than 200 arcsec. For the same growth direction, Okada et al. reported a FWHM of 211 arcsec [6].

2.2 Semipolar InGaN quantum wells

The improved quality of semipolar GaN with the help of a SiN interlayer and an additional cold top layer is now well-established and serves as a standard template recipe for subsequently deposited InGaN/GaN QW structures. By applying our standard quantum well growth conditions established for c-plane growth, five InGaN quantum wells at a temperature of about 775 °C were grown on a $(11\bar{2}2)$ template layer. The GaN barrier was deposited at a temperature of 835 °C. PL measurements show a significant difference to our c-plane reference sample grown in the same run: We measured a QW emission wavelength of 384 nm (cf. Fig. 6, left), whereas the (0001) QWs emit at 428 nm. By

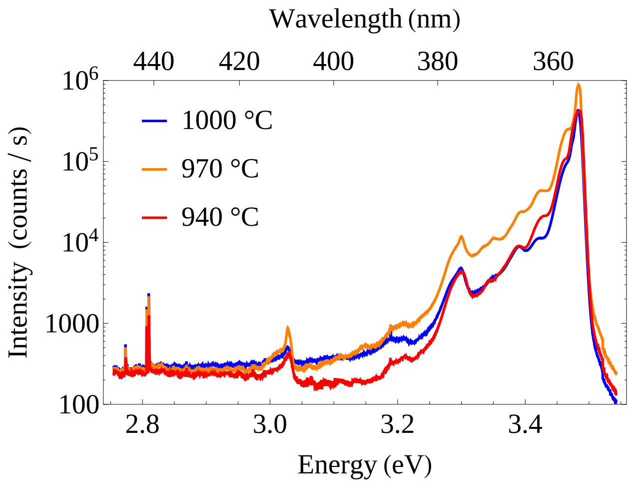


Fig. 5: Low-temperature PL spectra of $(11\bar{2}2)$ GaN with top layers grown at three different temperatures. For 970 °C, the GaN layer achieves an optimum crystal quality, indicated by the intensity of the D_0X emission at 3.482 eV.

evaluating the satellite peaks of HRXRD curves, we found indications for a significantly reduced QW periodicity in the case of the $(11\bar{2}2)$ sample (7.4 nm) compared to the c-plane counterpart (11.8 nm), translating into quantum well thicknesses of 2.5 and 4 nm, respectively. However, evaluations of the GaN growth rate at 1050 °C did not show such a difference (cf. Fig. 7). Hence, further investigations, e.g., TEM studies, should help to clarify this situation.

In the case of (0001) QWs, an emission wavelength of 428 nm is visible, whereas the $(11\bar{2}2)$ QWs emit at 384 nm (cf. Fig. 6, left). Measuring the quantum well dimensions via HRXRD shows a halved thickness in the case of $(11\bar{2}2)$ (2 nm) compared to c-plane (4 nm). Comparing the growth rate of (0001) and $(11\bar{2}2)$ GaN, it is conspicuous that they are exactly the same (cf. Fig. 7). TEM investigations should help to clarify this situation. However, in addition to the quantum well thickness, the piezoelectric field also plays an important role.

By doubling both, the indium flux and growth time of the $(11\bar{2}2)$ QWs, an emission wavelength of 424 nm was achieved (cf. Fig. 8, left), quite close to that of the c-plane reference sample. From the estimated InGaN layer thickness and the emission wavelength obtained on these samples, a rough estimation of their indium content is possible taking into account the different piezoelectric fields and quantization effects. In the case of the semipolar growth, an indium content of about 10 % is needed, to compensate the red-shift due to the QCSE in c-plane GaN (about 7 %). By increasing the indium flux from 53 µmol/min to 84 µmol/min, the emission wavelength could be further shifted to 486 nm, corresponding to an indium content of about 16 %. A sample with further increased quantum well thickness of about 6 nm emits at 500 nm. However, these green light emitting samples exhibit a strongly reduced quality, with just a weak signal of the quantum wells.

Similarly, quantum wells were grown on GaN. The InGaN layers were grown at a temperature of 735 °C and the GaN barriers at 800 °C. Using an indium flux of 42 µmol/min, a quantum well emission of 482 nm was achieved. By doubling the indium flux, the quantum

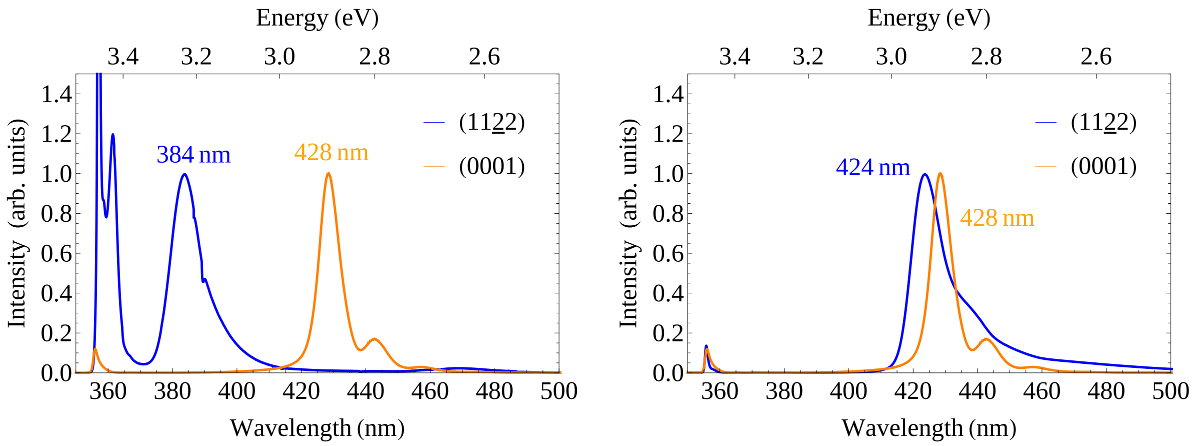


Fig. 6: Low-temperature PL measurements of (0001) (orange) and (11$\bar{2}$2) InGaN QWs (blue), respectively. When grown at comparable growth conditions, the semipolar QWs emit at shorter wavelength compared to the c-plane QWs (left). By doubling the growth time and the indium flux in the case of (11$\bar{2}$2) InGaN, the emission shifts from 384 nm to 424 nm (right).

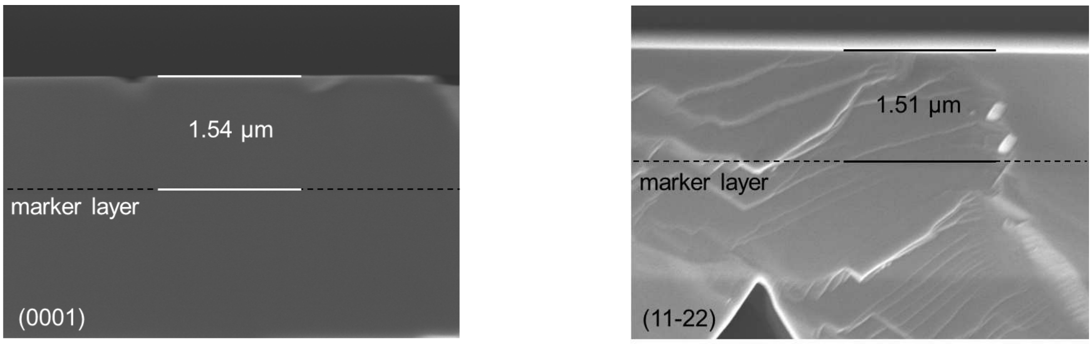
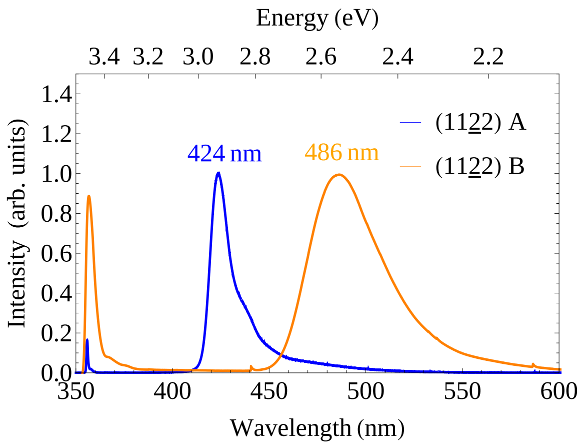


Fig. 7: Cross section SEM pictures of (0001) GaN (left) and (11$\bar{2}$2) GaN (right), respectively. Both samples were grown in one MOVPE run on different sapphire substrates. In both cases, we observed the same growth rate (within the limits of accuracy of the measurements).

well emission shifts into the green range to 525 nm. Obviously, the indium incorporation is significantly enhanced as compared to (11$\bar{2}$2), moreover, the quantum well quality on (10$\bar{1}$1) is much better, visible by a significantly higher PL intensity of the quantum wells. However, for the crystal quality of the pure GaN, we observed the opposite relation: the FWHM of the symmetric reflections of the (11$\bar{2}$2) rocking curves amount to less than 200 arcsec, whereas we found more than 400 arcsec for the respective (10$\bar{1}$1) sample.

A reduced quality of the (11$\bar{2}$2) InGaN quantum wells could be caused by a comparably rough surface morphology. We assume that an improved surface quality could help to improve the quantum well emission either. It turned out that the shape of the trench sidewalls has a big influence on the surface morphology. The sidewalls of the sapphire trenches, which were overgrown so far, has an angle of about 68 °(90 ° means perpendicular to the surface). However, the c-plane of the sapphire has an angle of 58 °. Changing the trench shape in such a way that the sidewall gets more c-plane-like shows very promising surface qualities.



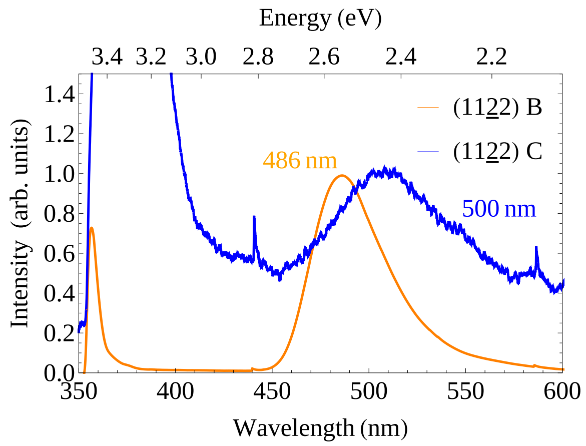


Fig. 8: Low temperature PL spectra of $(11\bar{2}2)$ InGaN quantum wells, grown at various temperatures with different thicknesses. Sample A has a QW thickness of about 4 nm and was grown with an indium flux of 53 μmol/min. Increasing the indium flux to 84 μmol/min increases the QW emission wavelength to 486 nm (sample B). In the case of sample C, the quantum well thickness was increased to 6 nm.

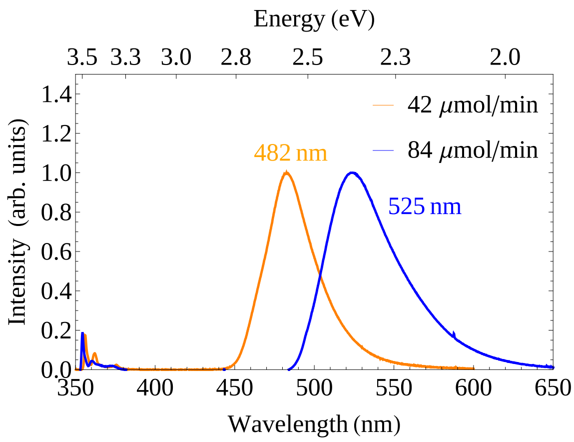


Fig. 9: Low temperature PL spectra of $(10\bar{1}1)$ InGaN quantum wells with different indium flux.

3. Conclusion

By inserting a SiN interlayer in the epitaxial process of $(11\bar{2}2)$ and $(10\bar{1}1)$ GaN grown on structured sapphire wafers, we were able to improve the quality of large area semipolar GaN. An additional top layer of GaN, grown at slightly lower temperatures leads to a pronounced overgrowth of the +c-wing over the −c-wing, hence blocking the defects in this area. By these techniques, the total defect density could be reduced significantly. On such optimized semipolar GaN layers, we have deposited InGaN quantum wells. However, when pushing the In content to larger values, just a weak quantum well photoluminescence emission at longer wavelength is detectable for $(11\bar{2}2)$ layers, whereas excellent properties have been measured on $(10\bar{1}1)$ templates with a strong QW emission at 525 nm.

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