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In-situ deposited SiN_x nanomask for crystal quality improvement in AlGaN

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We report the growth of high crystal quality $Al_{0.3}Ga_{0.7}N$ directly on sapphire substrates with metalorganic vapour phase epitaxy. We studied the improvements in crystal quality by introducing an *in-situ* deposited SiN_x interlayer. It acts as a nanomask which results in termination of the dislocations near the interface between the nanomask and epilayer. The epilayers with no SiN_x interlayer have very low density of screw type dislocations evident from transmission electron microscopy (TEM) investigations confirmed by a very narrow X-ray diffraction symmetric reflection (60”). This is a result of our oxygen doped AlN nucleation layer used to grow such epilayers. On the other hand, such epilayers have very broad XRD asymmetric reflections (typically a few thousand arcsec) indicating a high density of edge type dislocations. We could decrease the latter which are the main existing dislocations in our AlGaN epilayers. In TEM micrographs, we observed the formation of dislocation “bundles” when introducing SiN_x interlayers. This phenomenon promotes the dislocation free areas on the surface of the wafer. We carried out accurate optimizations of the SiN_x deposition. Consequently, we could grow high quality $Al_{0.3}Ga_{0.7}N$ epilayers with much narrower XRD asymmetric peaks—782” with total sample thickness of 1.4 µm.

1 Introduction Ultraviolet (UV) light emitting diodes (LEDs) are going to replace conventional bulky, toxic UV light sources bringing a lot of new advantages and applications such as air/water purification, UV curing and medical instrumentations. UV-LEDs and -laser diodes (LDs) are based on AlGaN. One of the most important factors in obtaining high optical output power is the crystal quality of the buffer layer. GaN, which can be grown with high quality, is not suitable here because most of the UV light will be absorbed in such a buffer layer. On the other hand, metalorganic vapour phase epitaxy (MOVPE) of AlGaN on sapphire typically leads to very poor crystal quality of subsequently grown layers if there is no modification of the growth technique. The high threading dislocation density (TDD) in such layers results in more non-radiative recombination of the carriers, consequently weakening luminescence from UV-LED or -LDs.

Epitaxial lateral overgrowth (ELO) of GaN has been well-established so far to improve crystal quality of GaN [1]; several groups have been successful in decreasing TDDs by applying ELO techniques in nano-scales with nano-masks – so called interlayers – like CrN [2], TiN [3] and more commonly used SiN_x [4]. The latter can be deposited *in-situ* during the growth. However, Engl *et al.* [5] could not observe any visible improvement in the crystal quality of $Al_{0.07}Ga_{0.93}N$ epilayers due to the SiN_x nanomask. It seems that the use of SiN_x interlayers for the improvement of AlGaN epilayers is a challenging issue. However, we could realize $Al_{0.2}Ga_{0.8}N$ epilayers with high crystalline quality (reduced dislocation density) just by a careful optimization of the SiN_x nanomask deposition process [6]. Light output power of UV-LEDs grown on those templates showed an increase by a factor of 20 in comparison to the UV-LEDs grown on the templates without SiN_x interlayers [6, 7, 8]. This motivated us to study the applicability of SiN_x nanomasks to realize a low number of dislocations – especially edge-type dislocations – in MOVPE grown AlGaN layers with higher Al-content (30%).

2 Experimental procedures All samples investigated in this study were grown on 2 inch (0001) sapphire substrates with a miscut of 0.3° towards the a-plane in a low-pressure horizontal reactor (Aixtron AIX-200/4 RF-S). Trimethylgallium (TMGa) and trimethylaluminum (TMAl) were used as group-III precursors, and ammonia as group-V precursor. The Al content in our samples was typically about 30% as confirmed by photoluminescence (PL). The process temperature was set to 1120 °C. As for our high quality GaN layers [4], we used a nucleation layer (NL) of oxygen doped AlN with a thickness of about 25 nm [9]. Silane was used for the in-situ deposition of the SiN_x interlayers with effective molar flow of 690 µmol/min [4]. The surface coverage of the SiN_x interlayers is controlled by the variation of the deposition time. High resolution X-ray diffraction (HRXRD) rocking curves (RCs) for crystal quality evaluation were performed on a Siemens D5000, equipped with a four bounce Ge (220) monochromator at the primary beam side in addition to a scintillation detector. In order to image the dislocations and identify different Burgers vectors **b**, the analysis of the TDs has been carried out by transmission electron microscopy (TEM) using the weak-beam dark-field (WBDF) technique by exploiting the **g * b** criterion, making use of the fact that $|\mathbf{g} * \mathbf{b}|^2$ is proportional to the intensity of the dislocation line in the image [1]. The WBDF images were recorded under the **g-3g** condition close to the [11-20] zone axis with a Philips CM-20 microscope. Root-mean-square (RMS) surface roughness extracted from atomic force microscope (AFM) images was evaluated on 10×10 µm^2 scans.

3 Results and discussion We have grown a reference $Al_{0.3}Ga_{0.7}N$ sample with a thickness of about 600 nm. HRXRD-RCs showed a very narrow symmetric (002) reflection (53”) and a very broad asymmetric (102) reflection (2590”). That means that despite a very low number of screw type TDs in the layers, there is a very high density of edge type TDs. In order to reduce the latter, which is more detrimental for device applications, we studied the use of SiN_x interlayers during the growth of $Al_{0.3}Ga_{0.7}N$. The SiN_x nanomask coverage can be readily controlled by changing the deposition time of SiN_x. The AlGaN growth conditions were not changed in all experiments done in this work - not even for overgrowth of the nanomask.

3.1 SiN_x nanomask after 150 nm In a first approach, we investigated structures where the *in-situ* deposition of SiN_x was done after 150 nm of $Al_{0.3}Ga_{0.7}N$ growth. After SiN_x deposition, again AlGaN was grown with a thickness of about 700 nm. We varied the SiN_x deposition time from 6 min to 9 min (see Table 1). Best result within this series were obtained for a deposition time of 6 min (sample: S3) yielding a HRXRD (102)-reflection with FWHM of 1406”. On the other hand, similar to our previous experiences with $Al_{0.2}Ga_{0.8}N$ [6], the (002) peak width increased with respect to the reference sample. Also here, we observed that the overgrowth of the SiN_x nanomask with AlGaN can lead to the formation of AlGaN nanoislands. In other words, the interlayer acts as an anti-surfactant [10] switching the growth mode from 2D to 3D growth mode. This can lead to an interrupted order in {0001} planes visible from broadened HRXRD symmetric reflections. Similarly, we could observe much rougher surfaces due to the formation of the AlGaN nanoislands. When continuing the growth to a final thickness of 1400 nm (sample M1), *in-situ* reflectance monitoring showed a completely recovered surface evident also from the smooth surface measured by AFM.

Table 1 The samples with a SiN_x interlayer deposited after 150 nm $Al_{0.3}Ga_{0.7}N$.

Sample	SiN (min)	(102) FWHM	(002) FWHM	RMS (nm)	Thickness (µm)
Ref.	0	2590”	53”	0.6	0.6
S1	6	1810”	172”	8.3	0.15+0.7
S2	7	1650”	228”	7.0	0.15+0.7
S3	8	1410”	285”	9.5	0.15+0.7
S4	9	1500”	350”	11.0	0.15+0.7
M1	8	1090”	217”	2.3	0.15+1.4

According to WBDF-TEM investigations, edge-type TDs (the main existing TDs) are stopped generally by the SiN_x interlayer (Fig. 1). TDs in some regions are merged or bent, creating bundles of dislocations, hence reaching the surface rarely separately. Obviously, this bundling increases the dislocation free surface effectively, in addition to dislocations annihilated by the SiN_x. However, the dislocation free areas on the surface are not as large as on our $Al_{0.2}Ga_{0.8}N$ [6]. As mentioned earlier, there is a very low number of screw type TDs in our AlGaN epilayers due to the oxygen doped-AlN NL. This has been confirmed by our TEM investigations (see Fig. 2). Detailed PL studies of such layers and TEM investigations which address the growth model and dislocation behaviour of such layers were published elsewhere [11, 12].

3.2 SiN_x nanomask on the NL As known from our previous studies [12], the composition of the epilayer below the SiN_x can influence the distribution of the nanomask. On the other hand, the position of the SiN_x may affect the crystal quality, similar to our experiences with $Al_{0.2}Ga_{0.8}N$ [6] and GaN [4]. Therefore, we have investigated the deposition of our SiN_x nanomask directly on the nucleation layer, based on sample M1 with a thickness of 1400 nm. Again, we varied the nanomask deposition time to establish the best surface coverage (see Table 2). Based on HRXRD-(102) reflections, sample N3 with 6 min deposition time shows a significant reduction of the edge-type dislocation density with a FWHM of 784”. As expected, the surface gets rougher with SiN_x deposition, accompanied by a broadening of the HRXRD-(002) reflections.

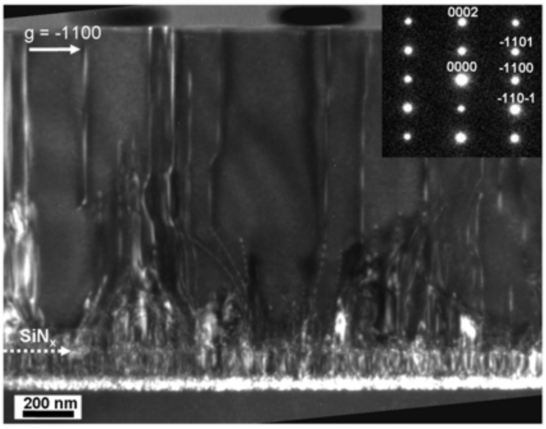

Figure 1 WBDF-TEM (g = -1100) micrograph of sample M1 with SiN_x interlayer 150 nm above the sapphire-AlGaN interface.

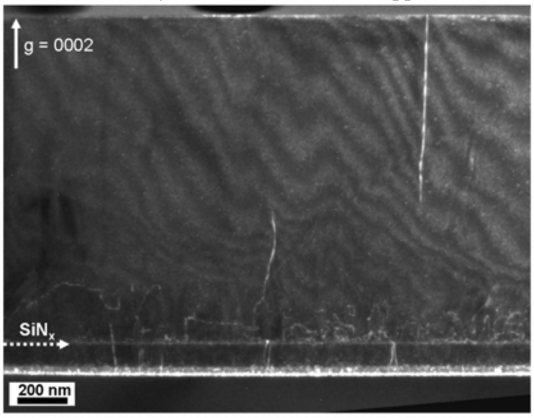

Figure 2 WBDF-TEM (g = 0002) micrograph of sample M1 confirming an extremely low density of screw type TDs.

Table 2 Samples with SiN_x interlayer deposited on the NL.

Sample	SiN (min)	(102) FWHM	(002) FWHM	RMS (nm)	Thickness (µm)
Ref.	0	2590”	53”	0.6	0.6
N1	4	1480”	324”	0.8	1.4
N2	5	1000”	338”	2.7	1.4
N3	6	784”	342”	1.3	1.4
N4	6.5	802”	259”	1.9	1.4
N5	7	857”	334”	2.6	1.4
N6	8	893”	336”	7.5	1.4

Obviously, a higher crystal quality could be achieved when the SiN_x nanomask was directly deposited on the NL in our $Al_{0.3}Ga_{0.7}N$ layers. This observation is contrary to our previous results for lower Al compositions — $Al_{0.2}Ga_{0.8}N$ [6] and GaN [4] where best results have been achieved for some distances between the SiN_x and the NL.

4 Conclusion The crystal quality of AlGaN epilayers can be improved dramatically with *in-situ* deposited SiN_x interlayers. Besides a fairly low density of screw type TDs in our layers presumably due to our oxygen doped AlN NL, we observed that a major part of the edge type TDs have been terminated at the in-situ deposited SiN_x interlayer. The remaining dislocations partly bend and form bundles, providing more defect free areas on the surface. The most effective position of the SiN_x nanomask was found to be on the NL. The SiN_x deposition time leading to best crystal quality is different for different positions of the interlayer. By carefully optimizing this deposition time, we decreased the width of the HRXRD (102)-reflection from 2590” in the reference sample to 784” in the sample with SiN_x deposited on the NL for 6 min demonstrating that such nanomasks can dramatically improve the crystal quality in AlGaN epilayers with at least 30% Al-content. Nevertheless, the effectiveness of *in-situ* SiN_x nanomasking in terms of crystal quality improvement seems to decrease with increasing Al-content in AlGaN epilayers.

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