MOVPE Growth of High Quality AlGaN Using In-Situ Deposited SiN Nano-Masks

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Growth optimization of AlGaN epilayers — with 20\% and 30\% Al content — directly grown on sapphire by MOVPE was investigated. Quality of the AlGaN epilayers was improved by an in-situ nano-masking employing ultra-thin SiN interlayer(s), confirmed by high intensity luminescence of quantum wells grown on top of these high quality AlGaN layers.

1. Introduction

AlGaN layers grown directly on sapphire typically experience fairly strong biaxial compressive strain [1] which leads to a large number of threading dislocations (TDs) — mainly misfit TDs. These mainly occur in the form of edge/mixed type TDs. They cause very broad asymmetric high resolution X-ray diffraction (HRXRD) reflections, e.g. (102)-reflexes [2]. On the other hand, screw/mixed type dislocations are mainly responsible for the broadening of the symmetric (002) HRXRD peaks. They do not hamper the device performance as strongly as edge type TDs [3]. Therefore, our main interest in this work is to grow AlGaN epilayers with as narrow as possible asymmetric XRD reflections corresponding to a low density of edge type TDs.

Nowadays, epitaxial lateral overgrowth (ELO) [4] techniques are widely used to grow high quality GaN layers using metalorganic vapor phase epitaxy (MOVPE). On the other hand, long ex-situ masking procedure and the presence of localized TDs in the window regions are typical drawbacks of ELO. Moreover, a thick overgrowth of the mask necessary to coalesce wing areas can lead to cracking of epilayers.

In-situ ELO techniques, especially in small scales, are of general interest due to the possibility to overcome the drawbacks of ELO while keeping its advantages. It has been shown by Tanaka et al [5] that SiN intermediate layers are an effective tool to grow high quality GaN epilayers. However, Engl et al. [6] observed that SiN interlayers do not reveal any visible improvement in crystal quality of AlGaN layers. In other words, these techniques seem to be less promising concerning the AlGaN material system, since AlGaN does not grow as selective as GaN [7]. Nevertheless, we report a successful implementation of SiN interlayers being deposited in-situ which could lead to facet-initiated ELO (FIELO) [8] in the nano-scale (nano-FIELO).

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2. Experimental

All samples investigated in this study were grown on (0001) sapphire substrates in a low-pressure horizontal reactor (Aixtron AIX-200/4 RF-S). Trimethylgallium (TMGa) and trimethylaluminum (TMAI) were used as group-III precursors, and ammonia as group-V precursor. The Al incorporation in our samples was typically about 20% and 30% as confirmed by photoluminescence (PL) and HRXRD. The standard growth temperature was set to 1120 °C. Similar as for our high quality GaN layers, we used a nucleation layer (NL) of oxygen doped AlN with a thickness of about 25nm [9]. Silane was used for the deposition of the SiN layers.

The thickness of the SiN interlayer must be less than a monolayer to obtain the best growth results [5]. An important factor in implementing an in-situ mask is the surface coverage, varied by the deposition time (Fig. 1).

In order to find the optimal coverage of the SiN mask, we deposited the mask after almost 150nm of AlGaN growth on the NL and grew nominally 1 μm above the mask. Also the effect of overgrowth thickness on the crystal quality has been investigated. A series of GaN-AlGaN multi quantum wells were grown on samples with different SiN modifications, taking their CL signal as a figure of merit in order to investigate the influence of the AlGaN layer improvements obtained by the SiN interlayers on device-relevant structures. These experiments were carried out similarly for samples with 20% and 30% aluminium.

As a reference for the following investigations, an AlGaN epilayer with 20% Al content was grown with a thickness of 0.7 μm (Fig. 2-a). The X-ray rocking curves (XRC) showed a very narrow symmetric (002) reflection — FWHM of 57" — and a very broad (102) asymmetric reflection — FWHM of 2350". The reference sample with 30% Al-content had a thickness of 0.6 μm showing 53" and 2590" XRC peak broadening for (002) and (102) reflections, respectively.
Phase-separation (inhomogeneous incorporation of Al in AlGaN epilayers) has a high probability to occur during AlGaN growth. The different surface migration lengths of the Ga and Al adatoms [1] play an important role in the phase-separation appearance, i.e. whenever growth occurs on an uneven surface, this phenomenon takes place more severely [10]. Since during AlGaN post-growth of SiN, pyramidal islands are initiated, we have investigated phase-separation as side effect by means of PL followed with locally resolved cathodoluminescence (CL), shortly called SEM-CL.

### 3. Results and Discussion

#### 3.1 AlGaN layers with 20% Al

AlGaN epilayers were grown as schematized in Fig. 2-b, with a SiN interlayer deposited after 150 nm growth of AlGaN on a NL followed by 1 μm overgrowth of SiN.

The deposition time of the SiN-layer was varied between 3 min and 8 min. The best results within the series could be obtained for a deposition time of 6 min revealing huge crystal improvement — XRC (102)-FWHM of 571° — due to SiN nano-masking, see table. 1. However, the more SiN coverage is, the rougher the surface, being evident from atomic force microscope (AFM) roughness evaluations in 10 μm × 10 μm. Whenever mask coverage is higher, consequently, more overgrowth thickness is essential in order to coalesce the initiated facets (submicro-islands), see Fig. 1. The surface roughness is also correlating with (002)-FWHM XRCs.

![Surface topography evaluations after different overgrowth of SiN mask (6 min deposited) with AlGaN.](image)

**Fig. 3:** Surface topography evaluations after different overgrowth of SiN mask (6 min deposited) with AlGaN.
Table 1: Effect of SiN deposition time on XRC (102), (002) reflections and surface roughness in AFM for the samples with 20% Al.

<table>
<thead>
<tr>
<th>SiN deposition (min)</th>
<th>(102)-FWHM (arcsec)</th>
<th>(002)-FWHM (arcsec)</th>
<th>RMS-AFM (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 min</td>
<td>2347</td>
<td>57</td>
<td>1</td>
</tr>
<tr>
<td>3:30 min</td>
<td>1789</td>
<td>115</td>
<td>2</td>
</tr>
<tr>
<td>4 min</td>
<td>1212</td>
<td>160</td>
<td>6</td>
</tr>
<tr>
<td>4:30 min</td>
<td>999</td>
<td>183</td>
<td>8</td>
</tr>
<tr>
<td>5 min</td>
<td>645</td>
<td>321</td>
<td>15</td>
</tr>
<tr>
<td>6 min</td>
<td>571</td>
<td>343</td>
<td>25</td>
</tr>
<tr>
<td>7 min</td>
<td>645</td>
<td>308</td>
<td>23</td>
</tr>
<tr>
<td>8 min</td>
<td>818</td>
<td>301</td>
<td>58</td>
</tr>
</tbody>
</table>

Therefore, we carried out a series of investigations concerning the effect of overgrown layer thickness after the SiN mask on crystal quality together with evolution of surface topography. The sample with a SiN deposition time of 6 min was chosen. Fig. 3 shows the surface topography and roughness evaluations done by AFM. After 2.5 μm overgrowth, we achieved sub-nm roughness values which was even less than our reference sample with 1 nm roughness. The asymmetric peak FWHM decreased with increasing thickness down to a value of 443" for the sample with 3.4 μm overgrowth (Fig. 4).

![Fig. 4: Effect of SiN mask overgrowth on XRC of (102) and (002) reflections in AlGaN (20%) epilayers — with SiN deposition time of 6 min.](image)

A multi quantum well (MQW) structure (8× GaN:3 nm/AlGaN:7 nm) was grown on these high quality layers to take their luminescence as a probe for the improvement of the epilayer quality due to the SiN interlayer. On the reference sample (AlGaN epilayer without SiN interlayer) the MQW-PL intensity at about 3.6 eV is weaker than that of the buffer layer — 700 nm thick AlGaN — at about 3.85 eV with PL-FWHM of 67 meV (Fig. 5). However, MQW grown on layers with a SiN interlayer at 150 nm and 1 μm overgrowth as buffer showed much more intense PL from MQW (PL-FWHM of 58 meV) compared to that of the barrier. The PL-FWHM of the MQW even became narrower when the SiN interlayer was directly deposited on the NL.

More complicated structures like deposition of two SiN interlayers with a distance of
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300 nm — the first one on the NL — even experienced narrower luminescence spectra with a FWHM of 42 meV.

According to weak beam dark field transmission electron microscopy (WBDF-TEM) investigations, edge-type TDs (the main existing TDs) are stopped generally by the SiN interlayer (Fig. 6-a). TDs in some regions are merged or bent, creating a bundle of dislocations reaching the surface. TDs can reach the surface but in the form of a bundle of dislocations, rarely separately. Thus, this bundling increases the dislocation free surface effectively, in addition to dislocations annihilated by the SiN. Fairly large defect-free areas with diameters in the range of few micrometers in lateral size could be observed (Fig. 6-a).

We interrupted the growth after nominally overgrowing about 290 nm AlGaN on the SiN interlayer in order to study TDs’ behavior due to SiN deposition. Scanning electron microscopy (SEM) of the surface topography reveals that some pyramidal islands are formed with \{1103\}-facets (Fig. 6-b). The islands look as if there is pure selective area grow of AlGaN on SiN nano-mask.

TEM cross sectional images from the same sample show that growth also occurs even between the formed islands(Fig. 6-c). The AlGaN grown on the wing area between the islands has a high density of TDs contrary to the pyramidal islands being almost TD free. The initiation of facets affect the TDs behavior as well. Due to the fact that growth in (1120) direction is faster than other in-plane directions the pyramidal islands have higher lateral growth rate. On the one hand, this leads to bending of TDs and even formation of dislocation loops, consequently dislocation termination. On the other hand, these high

Fig. 5: Normalized PL for reference sample (without SiN interlayer) and some high quality AlGaN layers with SiN interlayer. PL-FWHM of every layer is written on the respective graph.
quality pyramidal islands grow laterally in size and then dominate the low quality areas of AlGaN grown between them (on the wing area).

3.2 Phase separation investigations

The overgrowth of the SiN nano-mask was interrupted in order to record luminescence spectra of the topmost overgrown layers in different steps of the nano-FIELO (Fig. 7). PL spectra (Fig. 8) confirm that phase-separation phenomena occur, being evident from several visible luminescence peaks for each sample. At the very early stages of SiN-overgrowth, there are several peaks visible mainly in low Al-content regions. As the surface evolves - from sample ‘i’ to sample ‘v’ - two merging peaks are basically visible. With more overgrowth, consequently, there is just one main peak with a side peak, lower in energy as well as intensity.

In order to disclose how the Al (Ga) concentration is distributed over the surface, SEM-CL investigations were performed on the first sample within this series. The sample has 4 min AlGaN overgrown on the SiN mask deposited for 4 min.

In the CL spectrum of the sample (Fig. 9-a), several peaks are visible. The highest Al contents are incorporated on the top facets of the hillocks — c-plane facets evident from the 3.83 eV emission line (Fig. 9-e and Fig. 9-f). That is similar to findings of Zhoua et al. [11] that Al incorporated more into c-plane facets as compared to tilted m-plane facets. Together with the lower surface mobility of the Al-adatoms, there is another reason promoting phase segregation. Incorporation efficiency of Al and Ga adatoms depend also on crystallographic growth direction. The (001)-plane has a higher number of coordinated surface atom bonds than these tilted facets [12]. This means incorporation of adatoms on c-plane facets is energetically more favorable. In other words, Al-adatoms have stronger incorporation efficiency into c-plane facets due to lower surface diffusion length. This
results in a higher Al concentration on top of the hillocks and slightly less Al concentration in the side facets.

However, it is clear from SEM-CL mapping of the 3.46 eV emission line (Fig. 9-c) that pure GaN is also incorporated very much in the hillock areas. This can be attributed to the fact that in the beginning of the growth, as mentioned earlier, Ga has higher selectivity — less sticking coefficient — than Al. That results in diffusion of Ga adatoms to window area of the SiN mask at the very beginning of the AlGaN overgrowth while this is not the case for Al adatoms. Thus, it can be expected to observe GaN luminescence from the bottom layers of the hillocks. On the masked area, however, AlGaN growth is taking place (Fig. 6-c) but since it has mainly very poor crystal quality and less thickness than hillocks, it does not luminesce visibly.

### 3.3 AlGaN layers with 30% Al

Similar to our AlGaN (20%) layers, finding out the optimum coverage of SiN in AlGaN (30%) is our first step. Layer structures have been grown similar as described in Fig. 2-b with 1 μm overgrowth (OG) after SiN deposition. Obviously 8 min SiN-deposition time led to the best results (table 2), in the series with regard to the XRC (102)-FWHM being down to 1406” compared to that of the reference sample with 2591”. The same MQWs as in the previous part (AlGaN layer with 20% Al) were grown on these layers. We observed a huge improvement due to the SiN implementation into such layers with a PL-FWHM of 95.9 meV while that of the sample without SiN is 212.6 meV.
Fig. 8: PL spectra from the samples with different overgrowth times shown in Fig. 7.

Table 2: Effect of SiN-deposition time on XRD (102)- and (002)-reflections as well as on PL of MQW — emitting at 340 nm — grown on some of these buffer layers as a probe to enhancement of crystal quality in AlGaN (30%) layers.

<table>
<thead>
<tr>
<th>Structure</th>
<th>SiN (μm)</th>
<th>OG (min)</th>
<th>(102)-FWHM (°)</th>
<th>(002)-FWHM (°)</th>
<th>MQW PL-FWHM (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>i: SiN+1μm OG 0 min</td>
<td>2591</td>
<td>53</td>
<td>212.6 meV</td>
<td></td>
<td></td>
</tr>
<tr>
<td>i: SiN+1μm OG 6 min</td>
<td>1814</td>
<td>172</td>
<td>—</td>
<td></td>
<td></td>
</tr>
<tr>
<td>i: SiN+1μm OG 7 min</td>
<td>1652</td>
<td>228</td>
<td>—</td>
<td></td>
<td></td>
</tr>
<tr>
<td>i: SiN+1μm OG 8 min</td>
<td>1406</td>
<td>285</td>
<td>95.9 meV</td>
<td></td>
<td></td>
</tr>
<tr>
<td>i: SiN+1μm OG 9 min</td>
<td>1500</td>
<td>350</td>
<td>—</td>
<td></td>
<td></td>
</tr>
<tr>
<td>ii: SiN+2μm OG 8 min</td>
<td>1086</td>
<td>217</td>
<td>88.3 meV</td>
<td></td>
<td></td>
</tr>
<tr>
<td>iii: double-SiN 8 min</td>
<td>1087</td>
<td>233</td>
<td>74.5 meV</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

By continuing the AlGaN overgrowth on the SiN interlayers up to a thickness of 2 μm, we could further reduce the edge/mixed type TDs being evident from (102)-XRC, (sample ii in table 2). The PL-FWHM of MQW is also further reduced to 88 meV. A third sample (sample: iii in table 2) was grown with the same structure but with a second SiN interlayer with distance of 1 μm from the first SiN layer (Fig. 2-c). The second SiN did not lead to any change in the (102)-XRC peak width but revealed a slightly broader (002) reflection. Nevertheless, the MQW-PL was 13.5 meV narrower in the sample with double SiN interlayer.

4. Conclusion

In-situ deposited SiN nano-masking is an effective tool to reduce edge/mixed type dislocations in AlGaN layers. The dislocation reduction mechanism is basically FIELO in nano scale, leading to the realization of high quality layers without necessity of thick overgrowth of the mask. As AlGaN does not show a strong selectivity during overgrowth of SiN nano-mask, the growth occurs on the masked areas as well but the growth rate is
much lower than on the non-masked areas in the very beginning of the overgrowth of the mask, leading to the initiation of islands (facets) which have very low dislocation density. During the growth, they become larger eventually dominating the high dislocation density areas leading to formation of TDs’ bundle. The bundling effect additionally helps to realize larger dislocation free areas on the surface. However, this technique seems to be less efficient for higher Al concentrations, e.g. 30% Al content.

5. Acknowledgments

The TEM images provided by O. Klein and U. Kaiser from Central Facility of Electron Microscopy at Ulm University and AFM evaluations by R. Xue are gratefully acknowledged. This work was financially supported by the German Federal Ministry of Education and Research (BMBF) within the framework of the “Deep UV-LED” project.

References


