# Advanced PENDEOEPITAXY™ of GaN and Al<sub>x</sub>Ga<sub>1-x</sub>N Thin Films on SiC(0001) and Si(111) Substrates via Metalorganic Chemical Vapor Deposition

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# **Abstract**

Growth of GaN and  $Al_xGa_{1-x}N$  thin films on 6H-SiC(0001) and Si(111) substrates with low densities of defects using the PENDEO<sup>TM</sup> process and the characterization of the resulting materials are reported. The application of a mask on the GaN seed structures hinders the vertical propagation of threading dislocations of the seed material during regrowth, but introduces a misregistry in the overgrowing material resulting in low quality crystal growth. This misregistry has been eliminated due to advanced processing and the exclusion of the masking layer. The new generation of samples do not show any misregistry, as shown by transmission electron microscopy.

# Introduction

The PENDEO<sup>TM</sup> process is a new form of selective epitaxial growth that is dominated by the growth from sidewalls of rectangular stripes <sup>[1]-[5]</sup>. This process allows the growth of uniformly low defect density material over the entire surface of the semiconductor. Similar to LEO growth, a mask is employed to prevent vertical propagation of threading dislocations from the GaN seed structures into the regrown areas. The use of a mask can cause the formation of boundaries at the interface of coalescence of two growth fronts and a crystallographic tilt in the adjacent regions that have overgrown the mask. Electron-beam photolithography for the reduction in size of the GaN seed structures combined with the elimination of the masking material silicon nitride have been applied to achieve PENDEO<sup>TM</sup> growth of GaN and  $Al_xGa_{1-x}N$  on the aforementioned substrates showing no formation of coalescence boundaries and no tilt in the overgrown regions. Microstructural results via scanning electron microscopy and transmission electron microscopy, as well as X-ray diffraction spectra, have been obtained and will be discussed in the following sections.

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# **Experimental Procedure**

In the case of Si(111) as the substrate, a 3C-SiC buffer layer was deposited prior to any nitride growth, as described in Ref. [6]. On both substrates, 3C-SiC/Si(111) and 6H-SiC, films of GaN and  $Al_xGa_{1-x}N$  have been grown via the PENDEO<sup>TM</sup>-process on a 0.5 µm thick GaN(0001) seed layer grown on a high temperature, 100 nm thick AlN buffer layer. All nitride-based layers were grown using a cold-wall, vertical pancake style, RF-inductively heated metalorganic vapor phase epitaxy (MOVPE) system. The AlN buffer layers and the hexagonal GaN seed layers were each grown within the susceptor temperature ranges of 1080°C-1120°C and 980°C-1020°C, respectively, at a total pressure of 45 Torr. Triethylaluminum, triethylgallium, and NH<sub>3</sub> precursors were used in combination with a H<sub>2</sub> diluent. If employed, a 100 nm thick silicon nitride layer was used as a growth mask for blocking the continued threading dislocations during the PENDEO™ growth stage. The preparation of the GaN seed layer for this growth is explained in detail in Ref. [1]. Following these processing steps, the samples were degreased and cleaned prior to loading them into the MOVPE chamber. The PENDEO™ growth of the GaN and the Al<sub>x</sub>Ga<sub>1-x</sub>N layers were achieved within the susceptor temperature ranges of 1050-1100°C and 1080-1120°C, respectively, and at a total pressure of 45 Torr. Triethylgallium and NH<sub>3</sub> precursors were again used in combination with a H<sub>2</sub> diluent. The introduction of triethylaluminum into the growth chamber during PENDEO TM-growth produced Al<sub>x</sub>Ga<sub>1-x</sub>N layers having an atomic Al content of approximately 10%.

A JEOL 6400 FE scanning electron microscope (SEM) and a TOPCON 0002B, 200 KV, transmission electron microscope (TEM) were employed for microstructural analysis. A JEOL JAMP-30 high resolution scanning auger microprobe was utilized for measurements of the Al content of the as grown  $Al_xGa_{1-x}N$  layers.

### **Results and Discussion**

The first generation of GaN thin films grown by the PENDEO<sup>™</sup>-process are shown in Figure 1 for the substrate 6H-SiC and in Figure 2 for the substrate Si(111). A continuous film of fully coalesced GaN has been achieved by growing PENDEO<sup>™</sup>-GaN out of GaN seed posts covered with a silicon nitride mask to prevent vertical propagation of the threading dislocations of the seed post into the PENDEO<sup>™</sup>-GaN during regrowth.

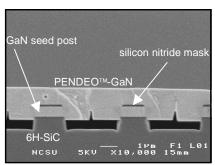


Figure 1. PENDEO™-GaN grown over masked GaN posts grown on a high temperature AlN buffer layer, on a 6H-SiC substrate.

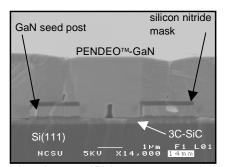


Figure 2. PENDEO™-GaN grown over masked GaN posts grown on a 2H-AlN/3C-SiC buffer layer, on a Si(111) substrate. A misregistry in the GaN above the mask is clear visible.

The SEM micrographs of Figure 1 and 2 indicate the influence of the silicon nitride masking layer on the quality of the PENDEO<sup>TM</sup>-GaN overgrowing the mask. Figure 1 shows void formation above the silicon nitride mask, as known from the LEO process<sup>[7]</sup>, which can be a result of a coalescence boundary formation, as presented in Ref. [8]. The formation of a coalescence boundary due to misregistry between the two growth fronts meeting above the silicon nitride mask is visible in Figure 2. Voids were being formed above the mask leading into a wavy surface morphology. The TEM micrograph shown in Figure 3 was prepared from a similar sample as presented in Figure 1. It shows the overgrowth region of PENDEO<sup>TM</sup>-GaN above the silicon nitride mask. The growth fronts coming from both sides of the masking layer are highly defective and form a coalescence boundary, which may act as an additional source for generating defects. Similar results are well know for LEO of GaN<sup>[8]</sup>. Additional to the negative impact of the masking layer in the quality of the overgrowing GaN, in the case of  $Al_xGa_{1-x}N$  the material tends to nucleate on the masking material, as presented in Figure 4 for  $Al_{10}Ga_{90}N$  using a silicon nitride mask.

100nm PENDEO™ GaN

Figure 3. TEM micrograph of PENDEO™\_GaN overgrown the silicon nitride mask. The coalescence boundary as a nucleation source for horizontal oriented dislocations is clear visible.

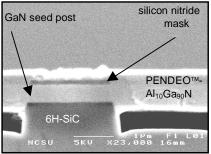
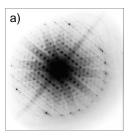


Figure 4. PENDEO<sup>TM</sup>-Al $_{10}$ Ga $_{90}$ N grown over a masked GaN post on a 6H-SiC substrate. The Al $_{10}$ Ga $_{90}$ N nucleated on the silicon nitride mask.

Subject of ongoing studies with lateral overgrown GaN films is the tilting behaviour. We conducted an XRD characterization of PENDEO<sup>TM</sup>-GaN grown from silicon nitride masked GaN seed posts on a 3C-SiC/Si(111) substrate. The rocking curves reveal one peak with a FWHM of 860 arcsec for a scan parallel to the posts and two peaks with a total FWHM of 2124 arcsec for a scan perpendicular to the posts<sup>[6]</sup>. This indicates that tilt is present in PENDEO<sup>TM</sup>-GaN grown over masked seed posts. But what regions of the PENDEO<sup>TM</sup>-GaN are responsible for the tilt? Small angle diffraction (SAD) pattern of coalescence regions of PENDEO<sup>TM</sup>-GaN above the trench between two seed posts, and above the masked seed posts of the later sample were prepared using TEM, as shown in Figure 5. The SAD pattern of the coalescence region above the trench between two masked seed posts does not show any evidence of two crystallographic orientations tilted

towards each other (see Figure 5(a)). In comparison, the SAD pattern of the coalescence region above the masking layer atop a GaN seed post shows clear evidence of the presence of two crystallographic orientations tilted towards each other (see Figure 5(b)). This behavior is also know from tilting studies of GaN grown via  $LEO^{[9],[10]}$ .



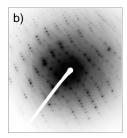


Figure 5. Small angle diffraction pattern of PENDEO™-GaN in the coalescence region (a) above the trench between two GaN seed posts and (b) above the silicon nitride mask atop a GaN seed post.

The application of a masking layer atop the GaN seed posts has the advantage of stopping the vertical propagation of threading dislocations of the seed post into the PENDEO™ regrown areas. But it also causes the formation of coalescence boundaries due to misregistry between the two coalescing growth fronts and tilt in the PENDEO™-GaN overgrowing the masking layer. Based on this facts a new generation of the PENDEO™-process has been developed. Electron beam (e-beam) lithography has been applied to reduce the width of the GaN seed posts into the submicron region and the masking layer has been eliminated. Figure 6 shows PENDEO™-GaN grown from submicron wide unmasked GaN seed posts on 6H-SiC substrate. No voids or misregistry above the seed posts are visible. The TEM micrograph in 6(b) shows a strong reduction in density of vertically oriented threading dislocations in the lateral and vertical grown PENDEO™-GaN compare to the seed post. Some dislocations were propagating vertically, but with reduced density. Noticeable is also that no dislocations were generated at the coalescence point of two growth fronts over the trench, above the void. SAD pattern from above the seed posts and above the voids did not show any evidence of tilt in this film.

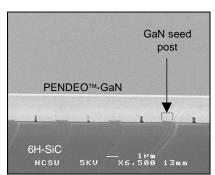


Figure 6(a). PENDEO<sup>TM</sup>-GaN grown on unmasked submicron wide posts on a 6H-SiC substrate. No misregistry above the GaN seed post is visible.

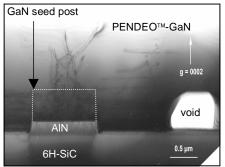


Figure 6(b). TEM micrograph of the sample shown in (a). A strong reduction in density of threading dislocations in the PENDEO™-GaN compare to the GaN seed post is seeable.

The same has been observed for PENDEO™-GaN grown from unmasked GaN seed posts on 3C-SiC/Si(111) substrates, as shown in Figure 7. The TEM image in 7(b) reveals a strong reduction in density of vertically oriented threading dislocations in the PENDEO™-GaN grown region. Some horizontally oriented dislocations were being generated at the GaN/AlN interface of the GaN seed post, which do not propagate to the surface of the film.

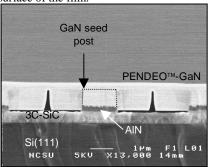


Figure 7(a). PENDEO™-GaN grown on unmasked GaN seed posts on a 3C-SiC/Si(111) substrate. No misregistry above the GaN seed post is visible.

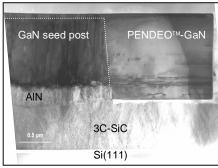


Figure 7(b). TEM micrograph of the sample shown in (a). A strong reduction in density of threading dislocations in the PENDEO<sup>TM</sup>-GaN compare to the GaN seed post is seeable.

Finally PENDEO<sup>TM</sup>-Al<sub>x</sub>Ga<sub>1-x</sub>N with Al contents of about 10% were grown on submicron wide, unmasked GaN seed posts on 6H-SiC substrates, as shown in Figure 8. The Al content has been determined by high resolution Auger electron spectroscopy and a large area scan revealed a variation in the Al content of +/-  $0.5\%^{[11]}$ . The elimination of the masking layer was necessary in this case after finding, that  $Al_xGa_{1-x}N$  nucleates on the silicon nitride masking layer, as observed in Figure 4. The TEM micrograph in Figure 8(b) shows no lateral propagation of vertically oriented threading dislocations of the GaN seed post into the PENDEO<sup>TM</sup>-Al<sub>10</sub>Ga<sub>90</sub>N regions. Only a reduced number of threading dislocations is propagating vertically out of the GaN seed post into the regrown areas. No dislocations are being generated at the meeting point of the coalesced growth fronts

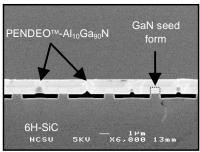


Figure 8(a). PENDEO™-Al<sub>10</sub>Ga<sub>90</sub>N grown on unmasked GaN seed posts on a 6H-SiC substrate. No misregistry above the GaN seed posts is observable.

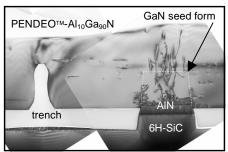


Figure 8(b). TEM micrograph of the sample shown in (a). A strong reduction in density of threading dislocations in the PENDEO™- Al<sub>10</sub>Ga<sub>90</sub>N compare to the GaN seed post is obvious.

above the trench of the PENDEO<sup>TM</sup>- $Al_{10}Ga_{90}N$  film. SAD patterns taken via TEM of the PENDEO<sup>TM</sup>- $Al_{10}Ga_{90}N$  film above the GaN seed post and in the coalescence region above the trench do not show any evidence of tilt.

#### Conclusions

Growth of GaN and  $Al_xGa_{1-x}N$  thin films on 6H-SiC(0001) and 3C-SiC/Si(111) substrates with low densities of defects using the PENDEO<sup>TM</sup> process have been reported. The disadvantages of applying a masked seed structure, namely formation of coalescence boundaries due to misregistry between the meeting growth fronts and tilt have been discussed and a new generation of the PENDEO<sup>TM</sup> process introduced. Overall, no evidence of coalescence boundaries and tilt were found in PENDEO<sup>TM</sup>-GaN and PENDEO<sup>TM</sup>-Al\_xGa<sub>1-x</sub>N grown films on 6H-SiC and 3C-SiC/Si(111) substrates using the new process. This is a different finding compare to similar grown structures on sapphire substrates, as presented in Ref. [12].

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