Mask-less epitaxial lateral overgrown MOVPE GaN layers on Si (111) substrates

S. Haffouz [1)], A. Grzegorczyk [2)], P.R. Hageman [2)], P. K. Larsen [2)], P. Vennéguès [3)] and E. W. J. M. van der Drift [4)]

[1)] Eindhoven University of Technology, Department of electrical Engineering, P.O.Box 513, NL-5600 MB Eindhoven, The Netherlands.

[2)] Experimental Solid Physics III, Research Institute for Materials, University of Nijmegen, Toernooiveld 1, 6525 ED Nijmegen, The Netherlands.

[3)] Centre de Recherche sur l'HeteroEpitaxie et ses Applications, Centre Nationale de la Recherche Sientifique, Rue Bernard Gregory, 6560, Valbonne, France.

[4)] Technical University Delft, Delfts instituut voor Micro-Electronica en Submicrontechnology, The Netherlands.

We report on the mask-less Epitaxial Lateral Overgrowth (ELO) of GaN on structured Si (111) substrates and on their structural properties using transmission electron microscopy (TEM), high resolution X-ray diffraction (HRXRD) and photo-electrochemical etching (PEC) techniques. TEM analysis shows that the overgrown regions only contain dislocations in the basal plane resulting from the bending of dislocations nucleated at the Si/AlN interface. The full width at half maximum (FWHM) in HRXRD of the rocking curve of the symmetric (0002) reflection is 30% lower in the GaN obtained by ELO process. The distribution of dislocations in the epitaxial layer is demonstrated by PEC technique.

Introduction

During the last few years, GaN-based devices have been demonstrated a large potential for applications in Opto-electronic domains. For all these applications, the III-V nitride layers are usually grown on sapphire or on silicon carbide substrates. However the development of some of these applications on silicon (Si) substrates has obvious technological advantages. The low cost, large-scale availability, good thermal and electrical conductivities and the feasibility of removing the Si substrates with wet etching are the main motivations for optimising the growth of III-Nitride material on this substrate. Due to the difficulties in the growth of GaN on silicon, caused by a lattice mismatch of 17% and by thermal expansion coefficient incompatibility, only few research efforts have been made to find out the successful growth process of GaN materials on such substrate. The standard growth process of GaN on Si (111), which uses only a low-temperature AlN buffer layer, still results in a high density of dislocations [1]. The use of multiple buffer layers [2] or a SiN_x [3] intermediate layer improved the optical and structural quality of the overgrown GaN films but the dislocations density is still as high as a few 10^9 cm^{-2}. However, as recently reported by few research groups [4-5], an improvement of the physical quality of GaN layers grown by Metalorganic Vapor Phase Epitaxy (MOVPE) can be achieved using the epitaxial lateral overgrowth (ELO) technique via selective mask. Although this technique has been proven to be very efficient to reduce the density of dislocations down to $5x10^7 cm^{-2}$ [5] using silicon substrates, it still needs a multiple regrowth steps procedure and *ex-situ* processing steps.

In the present paper we will report on the mask-less and single-step epitaxial lateral overgrowth of GaN on Si (111) substrates by MOVPE technique [6].

Growth procedure

The GaN epilayers were deposited on structured (111)-oriented silicon substrates inside commercial Metalorganic Vapor Phase Epitaxy (MOVPE) horizontal reactor (AIX.200). Trimethlgallium (TMGa), Trimethyaluminium (TMAl) and ammonia (NH_3) were used as the precursors of gallium, aluminum and nitrogen, respectively. The structured silicon substrates were prepared by conventional photolithography and dry etching techniques. The formed pattern consists of 4 µm-deep holes of 1.5 µm in diameter, each separated by 2.5 µm as shown in figure 1.

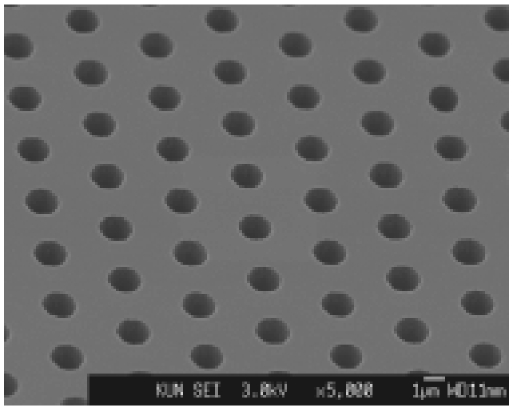

Fig. 1. Top view Scanning electron microscope (SEM) image of the structured Si (111) substrate.

Before loading into the MOVPE-reactor, the silicon substrates were first degreased in organic solvents, etched in a buffered 10% HF solution, and spin-dried. After loading, first an *in-situ* thermal cleaning procedure was applied to the surface during 10 min at 1100 °C in a hydrogen atmosphere followed by deposition of an aluminum nitride (AlN) nucleation layer at 850°C using 20 µmol/min TMAl and $4.1x10^{-2}$ mol/min NH_3. The optimum thickness of AlN buffer layer was about 10nm. Finally, the sample-holder is heated-up to a temperature of about 1170 °C for deposition of the gallium nitride (GaN) epitaxial layer.

Figure 2 (a) shows a scanning electron microscopy image of a 2µm-thick GaN layer. Clearly observable is that the deposition starts first between the holes, i.e. on the surface of the AlN film; no deposits are formed in the holes. These holes are intentionally deep etched in order to avoid nuclei-formation at the ridges or at the bottom of these structures. For incomplete coalescence we observe pyramidal holes showing six side facets. The two-dimensional evolution of the deposition is schematised in figure 2(b). This anisotropy is well known in GaN, which refers to the fact that the vertical extension (in <0001>) is much faster than in lateral direction (in <10-11>) resulting in a pyramidal shape [7]. In this study, the surface to be overgrown is about 1.5µm-wide, which makes complete coalescence possible only after 3µm-thick GaN deposition and without changing the other growth conditions like III/V ratio, growth temperature, NH_3 flow and without introduction of impurities like Magnesium (Mg) in order to enhance the lateral extension [8]. Contrary to the work of Sano *et al.* [9], the impurity contamination from the thermal decomposition of silicon substrate was not observed, which explains here the complete coalescence.

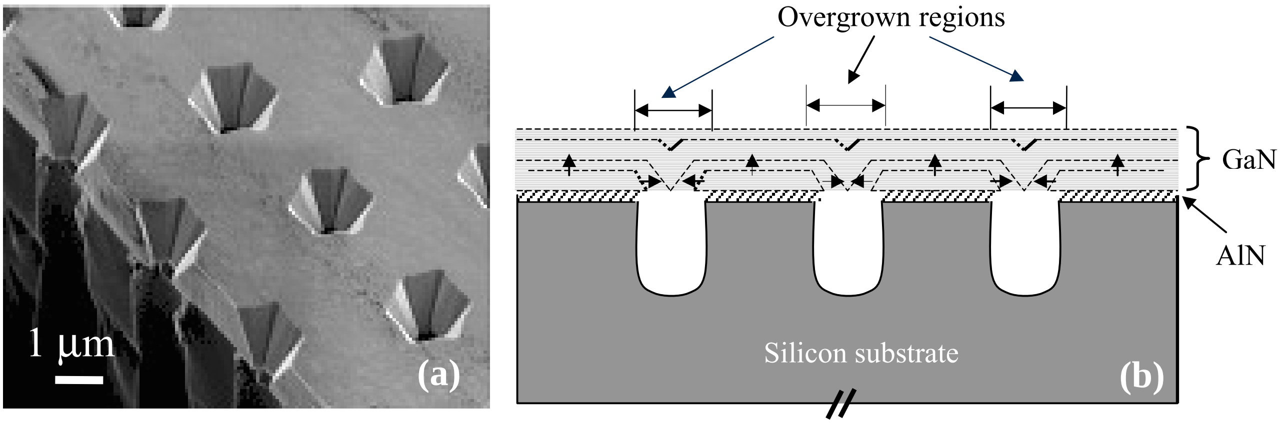

Fig. 2. (a) SEM image of 2µm-thick GaN layer and schematisation of the two-dimensional evolution of the deposition (b).

Structural characterization

During growth the GaN layer deposited between holes extends laterally and vertically over the holes until complete coalescence. This area has been closely studied by Transmission electron microscopy (TEM). TEM specimens have been prepared by a conventional combination of mechanical thinning and ion milling. TEM observations have been conducted on a JEOL 2010 FEG microscope. Figure 3 shows (0002) and (11-20) dark field cross-section TEM images of GaN laterally grown over the opening in the silicon substrate. Whereas only dislocations with a vertical line are observed above the Si substrate, the dislocations in the laterally overgrown regions are basal plane dislocations. It could clearly be seen that these dislocations result from the bending of vertical dislocations nucleated over the Si substrate close to the hole. This phenomena of the bending of dislocations is similar to what is observed in classical ELO growth. When a vertical dislocation encounters a lateral facet, it bends to adopt a direction in the basal plane. This mechanism is characteristic of lateral growth in GaN [10]. Most of the observed dislocations are a-type dislocations (Burger's vector = 1/3<11-20>). Some vertical dislocations are also observed in the coalescence boundaries between two overgrown regions.

High-resolution X-ray diffraction (HR-XRD) measurements showed a clear improvement of the structural quality of the GaN layer grown by ELO process, which reflected by the reduction of the full-width at half-maximum of the rocking curve of the symmetric (0002) reflection down to 594 arcs. Using standard process and for the same thickness of the GaN layer this value was 832 arcs [3].

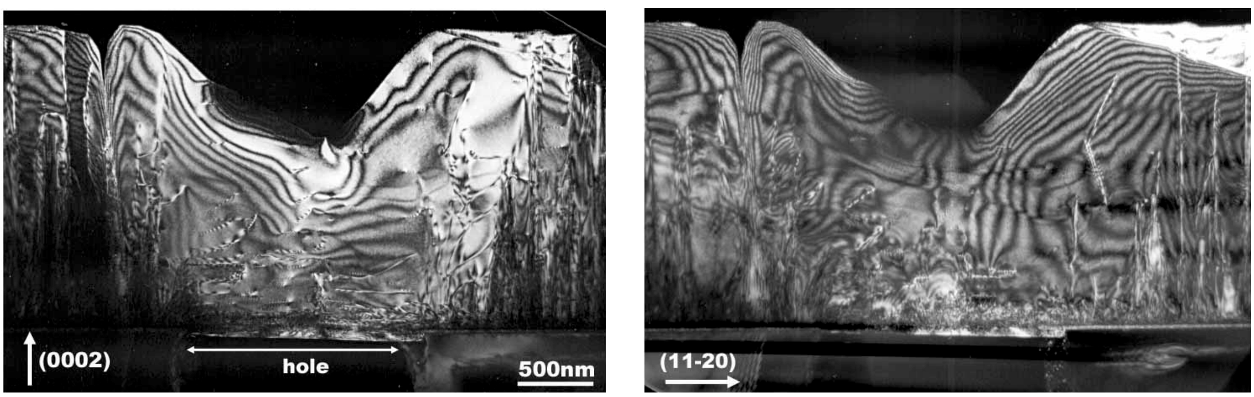

Fig. 3. (0002) and (11-20) dark field cross-section TEM images of GaN laterally grown over the opening in the silicon substrate.

Photo-electrochemical (PEC) etching was performed in a stirred KOH solution (0.004 molar) at room temperature. The UV illumination was provided by 450 W Xe lamp. A 100 nm-thick Ti layer was used to assure photocurrent conduction.
Figure 4 shows a SEM image of the GaN epitaxial layer after PEC etching in a stirred KOH solution (0.004 molar) at room temperature and under UV illumination. Straight whisker-like etch features are observed at the surface which reveals the distribution of dislocations in the epitaxial layer [11]. However these whiskers are formed only between the holes, i.e. not at the surface of the overgrown area. This result is consistent with what we observed by TEM i.e. no vertical dislocations are observed at the overgrown region. However the fact that PEC etching doesn't reveal the a-type dislocations need further and careful investigations.

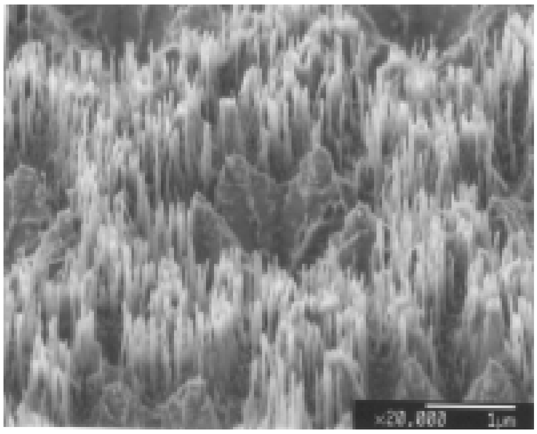

Fig. 4. SEM image of GaN epitaxial layer after PEC etching.

Conclusion

We reported on the mask-less and single step epitaxial lateral overgrowth of GaN by MOVPE technique using structured silicon substrates prepared by conventional photolithography and dry etching techniques. TEM analysis, HR-XRD measurement and PEC etching technique proved that this growth procedure is very promising for production of high quality GaN epitaxial films with very low dislocations density which is crucial need in order to improve the performance of opto-electronic devices based on these materials.

References

[1] A. Watanabe, T. Takeuchi, K. Hirosawa, H. Amano, K. Hiramatsu and I. Akasaki, J. Cryst. Growth, **128**, 391 (1993).
[2] E.Feltin, B. Beaumont, M. Laugt, P. de Mierry, P. Vennégués, M. Leroux and P. Gibart, Phys. Stat. Sol.(a), **188**, 531 (2001).
[3] P.R. Hageman, S. Haffouz, V. Kirilyuk, A. Grzegorczyk and P. K. Larsen, Phys. Stat. Sol.(a), **188**, 523 (2001).
[4] H. Marchand, N. Zhang, L. Zaho, Y. Golan, S. J. Rosner, G. Girolami, P. T. Fini, J. P. Ibbetson, S. Keller, S. Denbaars, J. S. Speck and U. K. Mishra, MRS internet J. nitride Semicond. Res. **4**, 2 (1999).
[5] E.Feltin, B. Beaumont, P. Vennégués, T. Riemann, J. Christen, J. P. Faurie and P. Gibart, Phys. Stat. Sol.(a), **188**, 531 (2001).
[6] S. Haffouz, A. Grzegorczyk, P.R. Hageman, P. Vennégués, P.K. Larsen, E. W. J. M. van der Drift, accepted for publication in Journal of Crystal Growth (2002).
[7] B. Beaumont, S. Haffouz and P. Gibart, Appl. Phys. Lett., **72**, 921 (1998)
[8] S. Haffouz, B. Beaumont and P. Gibart, MRS Internet J. Nitride Semicond. Res.**3**, 8 (1998).
[9] S. Sano, T. Detchprohm, S. Mochizuki, S. Kamiyami, H. Amano and I. Akasaki, J. Cryst. Growth, **235**, 129 (2002).
[10] P. Vennégués, B. Beaumont, V. Bousquet, M. Vaille, P. Gibart, J. Appl. Phys. 87 (2000) 4175.
[11] S. Haffouz, V. Kirilyuk, P.R. Hageman, L. Macht, J.L. Wehyer and P.K. Larsen, Appl. Phys. Lett, **79**, 2390 (2001).