Electron microscopy of AlGaN-based multilayers for UV laser devices

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Abstract. UV-Laser technology is one of the most challenging and important areas in nitride semiconductor research. A new growth technology, using a thin GaN interlayer approach, has recently been developed for UV-LEDs in our department. Improved optical quality of so grown LEDs has been demonstrated. A similar growth approach has now been used to grow an UV-laser structure that has shown stimulated emission (lasing) at ~340 nm via optical pumping. We report here a study of the microstructure, the epitaxial quality and the elemental distributions of such an aluminium gallium nitride-based heterostructure using a combination of various electron microscopy techniques.

1. Introduction
Ultraviolet (UV)-Laser technology is a very important area in nitride semiconductor research. GaN/AlGaN-based heterostructures are studied for various optoelectronic applications from biological agent detection to data storage and laser printing. The information density for these latter applications is defined by the laser spot area. This is limited by diffraction to the square of the emission wavelength [1]; hence the efforts towards producing shorter wavelength laser diodes (LDs) going to the deep-UV. Recently a laser device emitting at 342 nm has been demonstrated [2]. Conditions to achieve good emission properties are a good control of the elemental distribution during growth and a reduced density of crystal defects. We report here the structural and elemental analysis performed on a GaN-AlGaN multilayer, following a preliminary metal-organic chemical vapour deposition (MOCVD) growth project aiming to produce an UV-laser device emitting below 340 nm.

2. Sample description
Vertical low pressure MOCVD was used to grow the sample on α-alumina (sapphire). The initial thermal cleaning of the substrate under a molecular flux of hydrogen was followed by the deposition of a 0.5 μm thick AlN buffer layer at high temperature under group III-rich condition. A thin GaN interlayer (~20nm) was grown to reduce the dislocation density, followed by an Al0.75Ga0.25N/GaN superlattice used to prevent cracking. An undoped Al0.25Ga0.75N layer (~3.5μm) was then deposited followed by a silicon doped n-AlGaN cladding layer (~1μm), keeping the aluminium concentration fixed. Afterwards an n-Al0.15Ga0.85N guiding layer (100nm) was grown. The active region consisted of a multiple quantum well (MQW) structure with 3 Al0.05Ga0.95N thin layers separated by Al0.15Ga0.85N layers. On top of these, a p-type Al0.35Ga0.65N (15nm) blocking layer (to decrease electron flow), and then a p-type Al0.15Ga0.85N guiding layer (100 nm) and a p-type Al0.29Ga0.71N cladding layer (500 nm) were deposited. Finally, a 40 nm p-type GaN layer was grown to yield a good ohmic contact.

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3. Methods

A TEM cross-section sample was prepared gluing face to face two pieces of material. Grinding with SiC paper and final polishing with diamond powder was applied to the sample before Ar$^+$ ion milling to electron transparency. The analysis was performed on a JEOL 2010F TEM/STEM microscope operated at 197 keV and equipped with an Oxford Instrument Si:Li Pentafet EDX detector with ultra thin window and Link ISIS 300 acquisition software, annular dark field (ADF) (55 mrad inner angle) and bright field (BF) detectors coupled with a Gatan Digiscan system. A focused ion beam (FIB) AlN sample of (100±20) nm thickness was used for EDX and EELS calibration.

4. Results and Discussion

4.1. ADF imaging

The ADF image in Fig. 1a) shows the full device structure investigated in this work. The arrow points along the growth direction. The first dark band from the bottom represents the 0.5 µm AlN buffer layer. This appears darker because of its lower mean atomic number with respect to Al$_2$O$_3$ (substrate) and AlGaN (large middle layer). The sample appears to be crack-free, giving credit to the strain balance function of the Al$_{0.75}$Ga$_{0.25}$N/GaN superlattice. The various layers within the sample are planar and uniform except for the GaN interlayer that grows in form of islands as observed in [3]. The Al$_{0.75}$Ga$_{0.25}$N/GaN superlattice, as shown in Fig. 1b), covers the GaN islands. The contrast of the large cladding layer is quite uniform, except for some lines parallel to the growth direction ((0001) of wurtzite structure) that are due to misfit dislocations (MDs). Conversely, as shown in Fig. 1c), the guiding layer preceding the MQW was found to have a fluctuating Al concentration, resulting in a series of brighter and darker bands perpendicular to the growth direction that are visible from Z-contrast in ADF imaging. This fluctuation is also present in the p-type Al$_{0.15}$Ga$_{0.85}$N guiding layer deposited above the blocking layer. This variation is strongly correlated to the local Al concentration as discussed in section 4.3.

4.2. Thickness measurement by EELS

The sample thickness $t$ was estimated using Electron Energy Loss Spectroscopy (EELS). Low loss energy spectra were acquired over the energy window from -56eV to +456eV close to the position P5 in Fig. 1a) using a convergence angle of $\alpha=(10.0\pm0.5)$mrad and a collection angle of $\beta=(2.1\pm0.2)$mrad. The corresponding thickness was obtained via the Kramers-Kronig (K-K) sum rule [4] using the EELS routines of GATAN Digital Micrograph. This method was tested with an AlN FIB sample
(100±20)nm thick which gave a thickness of (150±5)nm and an elastic mean free path of \(\lambda=106\)nm. The results for the laser structure are reported in the Table 1 together with the position where the spectra were acquired. Starting from a thickness of 403nm near the AlN substrate, the thickness decreases towards the active layer to about 300nm but then increases at the blocking layer to 560nm. This thickness variation could have been caused by a preferential thinning rate during the ion milling process associated with an incidence angle of ~6 degrees with respect to the sample plane.

4.3. EDX analysis

A preliminary analysis of the AlN FIB reference sample was undertaken to test the ISIS 300 X-ray quantification routine [5] on a spectrum acquired under the same acquisition conditions as for the LD sample. By varying the nominal input thickness it was found that the expected ratio of 1:1 for Al:N was obtained only at \(t=0\) while for the true sample thickness \(t=150\)nm the apparent N concentration reported was 55at%. This indicates both, a relative error in the \(k\)-factor above 10% and a faulty calculation of the absorption. Lacking a reference sample containing Al, Ga and N at known ratios to be used for a calibration of the \(k\)-factors and the absorption coefficients, we have used for our analysis two approaches. A first quantification was obtained by using the ISIS software under thin film approximation (input \(t=0\)nm) and a second quantification by using an internal calibration of the \(k\)-factor assuming the nominal composition in the middle of the cladding layer (point P5 in Fig. 1a). This position is far enough from the substrate to limit the Al contribution from it. The \(k\)-factor was obtained using the Cliff-Lorimer (C-L) equation

\[
\frac{C_A}{C_Ga} = k_{A/Ga} \cdot \frac{I_A}{I_{Ga}},
\]

where \(C\) is the elemental concentration in atomic percent (at%), and \(I\) the area under the full-width-at-half-maximum of the peak of interest (background subtracted with the software ISIS). Only Al-K and Ga-L peaks were considered in order to have similar absorption for the changes in thickness across the sample. For the spectrum in position P5 we found a value of \(k_{A/Ga}=1.56±0.04\).

A series of EDX point analyses was performed along the whole length of the sample to asse the Al concentration in each layer at the positions indicated in Fig. 1 (see Table 1). The AlN buffer (P1) contains no Ga, as expected. The superlattice (P2) gives a value of \(x=0.65\), a value intermediate between its \(Al_{0.75}Ga_{0.25}\)N and GaN components. The interlayer islands in Fig. 1b) consist mainly of GaN; the 10at% Al fraction (P3) is probably a contribution from the surrounding superlattice partly covering them along the beam direction. The Al content is close to the nominal values in the various layers. Larger values than nominal are found for the Al-rich blocking layer and the quantum wells. The beam broadening, estimated using the ISIS software for a sample thickness of 500nm, is ~90 nm. This is much larger than the QW thickness (~4nm) and indicates there is a significant contribution from the neighbouring layers. The concentration fluctuations visible in the active area of the LD shown in Fig. 1c) are evident from the EDX data (P6-P7, P10-P11, P12-P13) where a darker area in ADF corresponds to a higher Al fraction in the material. More accurate quantification could be obtained from a thinner sample in order to reduce the beam broadening; also more thickness uniformity would improve the method of internal calibration.
4.4. Two-beam analysis
Dark film images obtained in two beam conditions with \( g=(11-22) \) are shown in Fig. 2a,b). Along this orientation all dislocation components are visible. A high density of MDs is generated, as expected, at the interface between the sapphire substrate and the AlN buffer. The GaN interlayer layer was found to filter the number of dislocations propagating to the undoped \( \text{Al}_{0.29}\text{Ga}_{0.71}\text{N} \) layer, as in [3]. Some of the MDs form a dense network, as indicated by arrows in Fig. 2a), others propagate towards the active region of the device passing through the MQW, blocking layer and reaching the top GaN layer (see Fig. 2b). In addition, dislocations are also formed directly within the guiding layer and MQW active region, as shown in the ADF image of Fig. 2c). The overall good material quality of this heterostructure was confirmed by the stimulated emission (not reported here) at \( \sim 340 \) nm wavelength, obtained via optical pumping based on a control sample.

![Figure 2. TEM DF images obtained selecting two beam conditions with exciting vector \( g=(11-22) \), near the buffer layer (a) and the LD active region (b). c) ADF image showing dislocations generated in the MQW, from the area C in Fig. 1a)](image)

5. Conclusions
A GaN-AlGaN laser diode structure showing stimulated emission at \( \sim 340 \) nm via optical pumping on a control sample was investigated using electron microscopy imaging and analytical techniques. Elemental concentrations in the multilayers were found in agreement with the nominal device structure within the limit of the techniques used for the analysis. An additional unwanted concentration modulation was revealed by Z-contrast imaging and EDX analysis. Misfit dislocations, reduced in density by the GaN interlayer, were found to propagate through the cladding layer up to the top layer together with dislocations generated in the active region.

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