Standard Project

Experimental Report template

Proposal title: **Strain in AlGaN/GaN nanostructures**		Proposal number: 20150512 32-02 783
Beamline: BM32	**Date(s) of experiment:** from: to:	**Date of report:** 17/09/2016
Shifts: 15	**Local contact(s):** Jean-Sébastien Micha, Samuel Tardif, François Rieutord	*Date of submission:*

Objective & expected results (less than 10 lines):
Determination of strain in new MOCVD aluminium nitride nanostructures within the pillar geometry. Use of grazing incidence X-ray diffraction to determinethe strain tensor. Correlation with Aluminium composition and optical properties.

Results and the conclusions of the study (main part):

In this study, high-quality and thin III-nitride tubes having a wall only composed of GaN/InAlN multiple quantum wells (MQWs) are fabricated by a simple full nitride metal-organic vapor-phase epitaxy approach. The synthesis of such MQW-tubes is based on the c-axis vertical GaN wire growth surrounded with core-shell MQWs heterostructure followed by a selective etching using *in situ* controlled H_2/NH_3 annealing to remove the inner GaN wire (See Figure 1). Well-defined MQW-based tubes having non-polar *m*-plane orientation exhibit significant UV light emission near 330 nm up to the room temperature. The strain involved in non-etched and etched (tubes) structures have been measured by GIXRD and measurements of truncation rods.

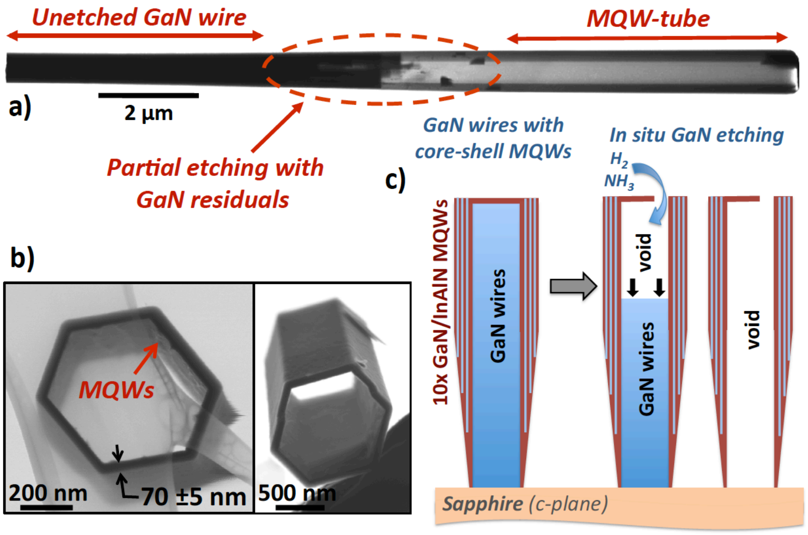

Figure 1: Formation of MQW-tubes after an uncompleted wire etching by the in situ H_2/NH_3 annealing of GaN wires coated with radial 10x GaN/InAlN MQWs. a,b) STEM images at 20 kV of dispersed MQW-tubes on holey carbon membrane having a tube wall thicknesses of 70 nm corresponding to the expected thickness of 10 MQWs. c) Schematic of the etch mechanism by the in situ H_2/NH_3 annealing in the case of thick MQW shell.

As already shown by our group, GaN wire templates are in epitaxy on the sapphire substrate and Grazing incidence X-ray diffraction (GIXRD) has been used to confirm the in-plane 30° rotation between the two unit cells, *i.e.* $[1\,0\,\bar{1}\,0]_{GaN} \parallel [1\,1\,\bar{2}\,0]_{Sap.}$ and $[0\,0\,0\,\bar{1}]_{GaN} \parallel [0\,0\,0\,1]_{Sap.}$. Experiments have been performed on as-grown and etched GaN/InAlN core-shell samples with 9.5 keV photon energy (λ=0.13051 nm) at the BM32 beamline of the European Synchrotron (ESRF in Grenoble). A sample with bare GaN wires has also been measured to provide a reference. The grazing incidence angle is set to 0.15 °, *i.e.* lower than the critical angle of refraction (around 0.24° at this energy) to strongly enhanced the intensity diffracted by the wires/tubes with respect to the substrate surface. The samples are aligned with respect to the usual hexagonal c-plane sapphire surface unit cell (a=0.4758 nm, c=1.2991 nm) to define an orientation matrix that uses $(HKL)_{Saph.}$ Miller indexes and sapphire reciprocal lattice (rlu). This choice allows accurate scanning of the reciprocal space. Figure 2 shows in-plane diffraction scans along the a) $[H\ 0\ 0]_{Sap.}$ direction exhibiting the $(H\ 0\ 0)_{GaN}$ reflection due to epitaxial relationship and b) $[H\ H\ 0]_{Sap.}$ direction to measure the $(H\ 0\ 0)_{GaN}$ reflection (see larger scans showing sapphire peaks in supplementary materials). The in-plane interatomic distances corresponding to the usual (110) and (200) GaN interplanar distances can be measured in the three samples. They correspond to the GaN core, even for the “tube sample”, which is not completely etched. Moreover, an additional peak is measured at larger rlu that we attribute to the GaN/AlGaN shell and more precisely to the 0-order peak of the MQW.

The peak broadening of the core-shell “wire” sample (non etched and wit MQW) is wider and probably correspond to a mixing of different contributions including AlGaN substrate overgrowth. The etching of the wires provides a narrower peak with a stronger contrast clearly defined both for the (110) and (200) reflections (see arrows). From the direct measurement of the peak positions with respect to the GaN internal reference, we can deduce a decrease of the average interplanar distances by about -0.44 % for these two plane families suggesting a smaller InAlN lattice parameter than bulk GaN. The interpretation in terms of composition inside the InAlN layer is difficult because it must take into account the nature of the MQW (thickness and composition) as well as anisotropic strain and relaxation. This work will be focused in another studies. Nevertheless, this diffraction result suggests that the GaN quantum well layer inside the InAlN/GaN MQW of the “tube sample” should be in tension to balance the average strain corresponding to the average peak. As also shown in supplementary materials, grazing incidence angle measurements of crystal truncation rods along the l direction for (h,k) values indicated by #1,2,3,4 in Fig. 2 have confirmed a smaller value of the average lattice parameter along the –c growth direction with a decrease of -1.15 % (resp. -1.3 %) for the measurements along (1 1 l) for the “tube” and “wire” samples.

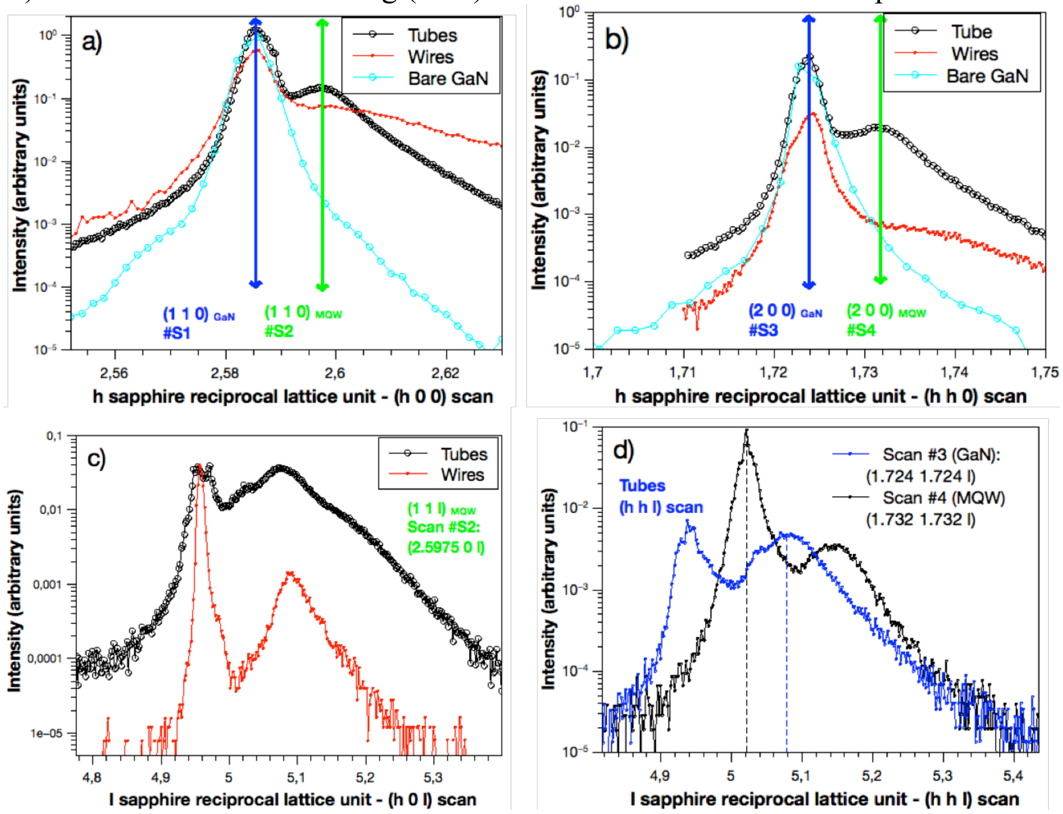

Figure 2: Grazing incidence X-ray diffraction of assemblies of wires grown on c-sapphire substrate: bare GaN wires, GaN wires coated by core-shell GaN/InAlN multiple quantum wells (called “Wires”) and “Tubes” obtained by H_2-etching the core of the previous sample. Reciprocal lattice units correspond to the c-sapphire substrate lattice and the a) $(110)_{GaN}$ and b) $(200)_{GaN}$ peaks are shown. Positions indicated by #1,2,3,4 can be used to perform out-of-plane measurements at given (h,k) positions.

These recults motivate also our group to perform finite element calculations to simulate the general tendancies of this new type of relaxation by changing the relative quantum well and quantum barrier thicknesses.

Justification and comments about the use of beam time (5 lines max.):

The possibility to simply fabricate thin nitride tubes with embedded MQW-based active region associated to controllable optical emission properties opens a way to develop nitride tube-based device applications.
All the objectives and expected results have been fulfilled by our experiments including the understanding of optical properties features of these objects (see Fig. 3).

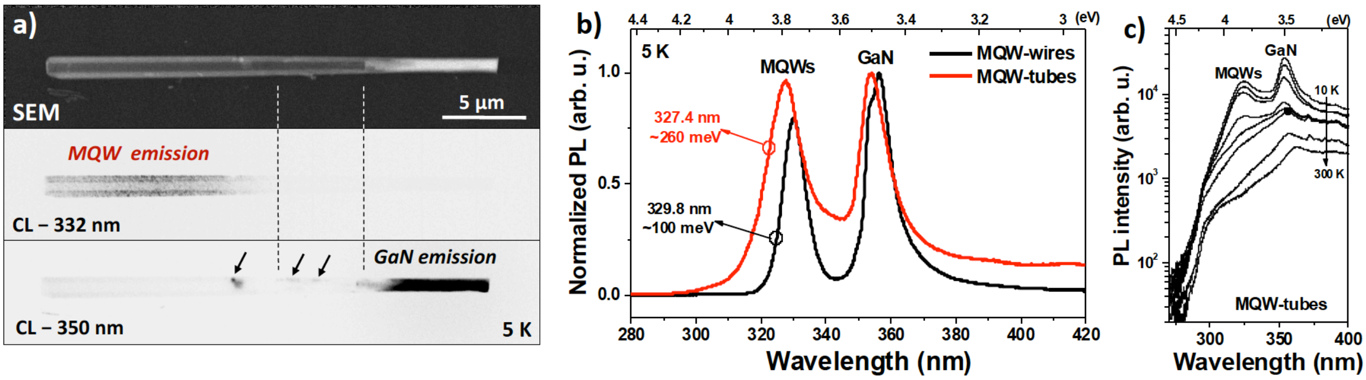

Figure 3: Optical light emission properties of MQW-tubes based on CL and PL measurements. a) SEM and CL mapping at 332 and 350 nm at 5K measured on typical single MQW-tubes (light emission corresponds to dark regions). b) Comparison of PL spectra at 5K measured on ensemble of dispersed wires before and after the in situ etching annealing labeled as MQW-wire (black curve) and MQW-tubes (red curve), respectively. c) Temperature-dependent PL measurements from 10 to 300 K measured on dispersed MQW-tubes ensemble.

Publication(s):
- These results will be submitted this year (before the end of 2016) most probably in ACS Photonics.