Understanding the MBE growth of GaN nanowires with in-situ grazing incidence diffraction

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III-Nitride semiconductor material is of major importance for blue and UV light emission applications. Unfortunately, the potentialities of the material are still plagued by the high density of dislocations (10^{10} cm^{-2}) and other structural defects which result from the lack of lattice-adapted substrates. GaN nanowires grown by MBE exhibit an exceptional crystalline quality and could be an option to overcome this difficulty. These defect-free nanostructures motivate a serious hope for creating high efficiency III-V nanowires based devices.The nucleation process of nitride nanowires (NWs) grown by molecular beam epitaxy (MBE) remains unclear. It was the aim of this in-situ diffraction experiment, in grazing incidence, to elucidate the nucleation process. For instance, what are the seeds/precursors for NWs growth ? How are they forming on the surface (Stransky-Krastanov process or Volmer-Weber one) ?

The samples were grown in situ in the SUV chamber. First, a thin (3nm) AlN buffer was grown on a Si(111) substrate. The AlN buffer enhance the orientation and the homogeneity (size, high) of the wires. Then at a high substrate temperature (760°C) (with respect to the temperature growth of conventional nitride layer or quantum dots) we exposed the surface with Ga and N fluxes with a Ga/N ratio lower than 1 (N rich condition). Once optimized, this growth conditions gave rise to GaN nanowires well aligned along the c-axis.

The diffraction experiment consited in measuring the in-plane 30-30 Bragg reflection of the GaN seeds/NWs and AlN buffer continuously during the growth. Grazing incidence setup, with an incident angle equal to 0,2° for the sample grown with the medium Ga flux and 0,15° for the samples grown with the higher and lower Ga flux, allowed us to be more sensitive to phenomena occuring on the surface by drastically decreasing the Si scattering. One advantage to work at synchrotron (ESRF) is the amount of photon available permiting us to decrease the scan duration at about 15 seconds and still having a good noise/signal ratio. The shorter is the time scan higher is the sensitivity to the very early stage of the growth. Furthermore we decided to reduce the amount of Ga impinging on the surface and hence slow down the nucleation process. We grew three sample with the same substrate temperature and N flux but with differents Ga fluxes. Fig 1 shows *h*-scans across GaN and AlN 30-30 Bragg peaks taken a different growth time for the sample grown with the medium Ga flux. Figure 2 shows the integreted intensity of the GaN diffraction as a function of time for the 3 different Ga fluxes.

These experiments brought about two main results:

1. Decreasing the amount of Ga impinging per second on the sample surface results in delaying the 3D growth.
2. Once the growth has sarted, the GaN diffracted intensity is a quadratic function of time at the beginning of growth then it is linear.

These results suggest three different stages during the GaN NWs formation :

1. First of all the growth begin with the formation of a layer of dots on the thin AlN buffer by a Stransky-Krastanov process. According to this process the formation of a two dimentional (2D) layer is followed by a three dimentional (3D) growth mode (dots formation). We interpret the delay to the nucleation as being the time for the formation of this 2D layer. Note that the substrate temperature is high, then the Ga sticking coefficient is low, this explain why it takes such a long time to deposit only 2 ML of GaN.

2. Once the dots layer is formed, the sticking coefficient of Ga adatom increases. Indeed the dots offer new nucleation centers where Ga and N adatoms can be adsorbed. Therefore the growth speed increases and enter in a coarsening stage. At the early stage of growth, the GaN diffracted intensity can be fitted with the equation A_0*t^x where A_0 and x were adjusted. We have found an exponent close to two for the three samples, this depict a coarsening by the sides of the dots.

3. And finally the dots end to grow lateraly as a consequence they develop only by their top and give rise to the NWs growth. This explain the linear behavior of the growth after the quadratic one.

These growth behavior: high lateral/low vertical growth rate for the dots at the early stage followed by a high vertical/low lateral growth rate for the wires has been first suggested Songmuang and *al.* (Appl. Phys. Lett. **91**, (2007), 251902). Our x rays diffraction results strongly support this model of growth.

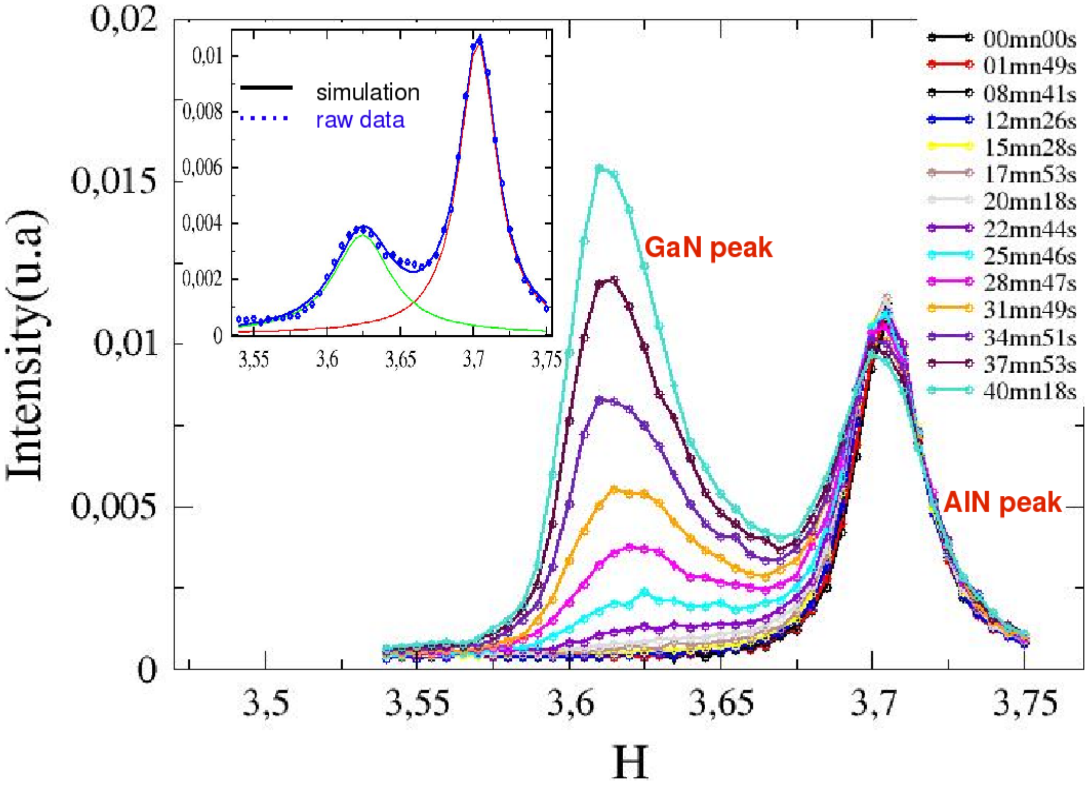

Figure 1: Bragg peaks of the 30-30 GaN precursors/NWs (3,62) and AlN buffer (3,7) recorded as a function of time time for the sample with the medium Ga flux. The incident angle was equal to 0,20°. The inset shows the experimental and fitted h-scans recorded after 31 minutes of growth. This fit is made of two pseudo-Voight function : one corresponding to the AlN peak and the other to the GaN peak.

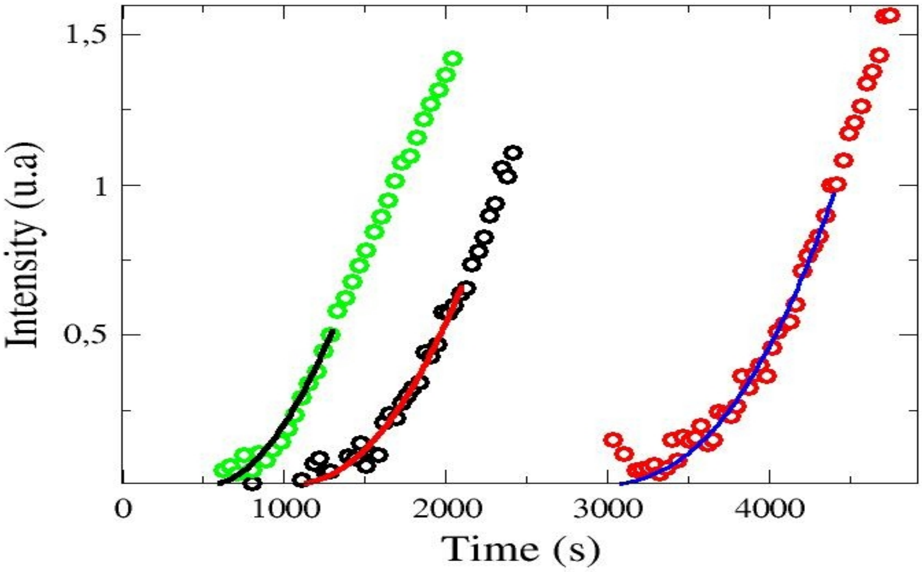

Figure 2: Open symbols represent the integrated area under the GaN peak for the three sample grown with differents Ga flux. Continuous lines are fits with the equation A_0*t^x where A_0 and x were adjusted. Two effects are induced by decreasing the Ga flux : the time delay to the growth occurrence increases and the rippening is longer. Note that only the rippening is influenced by the Ga flux, after reaching the linear growth mode the growth rate is the same for all samples. This fact point out the fundamental role of the Ga adatom diffusion occuring during the NWs growth (see also Songmuang and *al*., Appl. Phys. Lett. **91**, (2007), 251902.