Structure of GaN quantum dots grown on AlN by plasma assisted molecular beam epitaxy by means of grazing incidence synchrotron radiation

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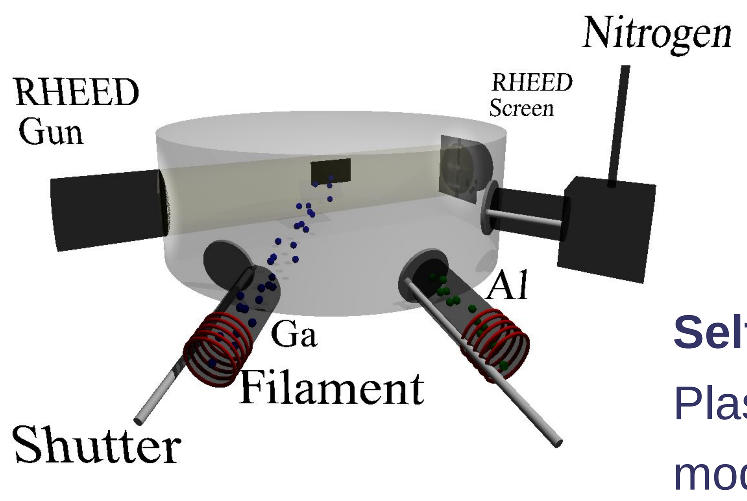
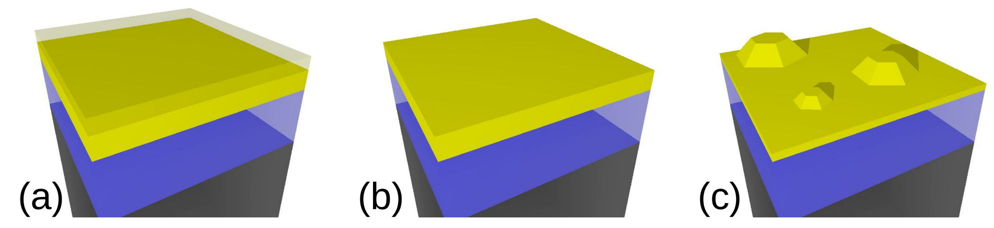
Molecular beam epitaxy growth

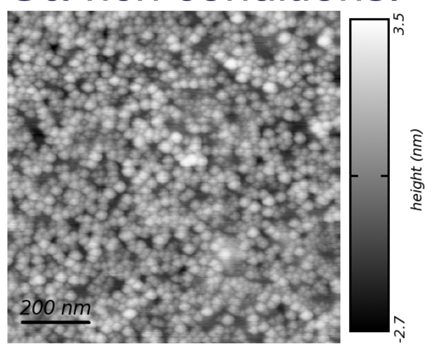




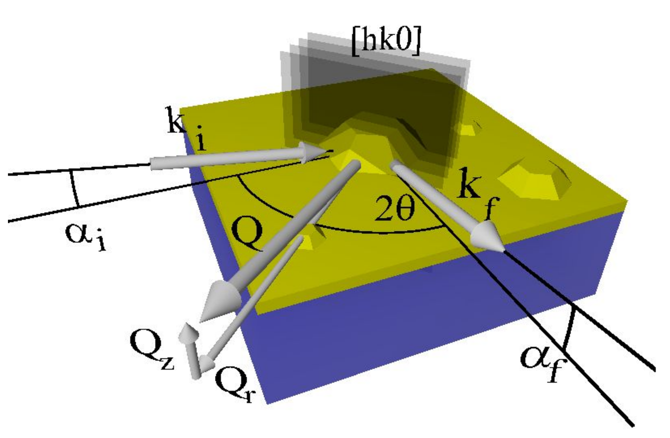
Self-organized quantum dots (QDs) are grown by Plasma Assisted Molecular Beam Epitaxy in the modified Stranski-Krastanow (SK) mode[1].

The growth process falls into 3 steps : (a) a 2D GaN is grown in Ga-rich conditions. The SK transition is inhibited by a Ga layer. (b) the Ga layer is evaporated under vacuum. (c) 2D/3D transition followed by a ripening process. 6H-SiC or AlN (0001) substrates are used.

⇒ diameter ~ **20 nm**, height ~ **5 nm**,
density control range **[2.10^{10}-2.10^{11}] QDs/cm^2**



Grazing Incidence Multi-wavelength Anomalous Diffraction

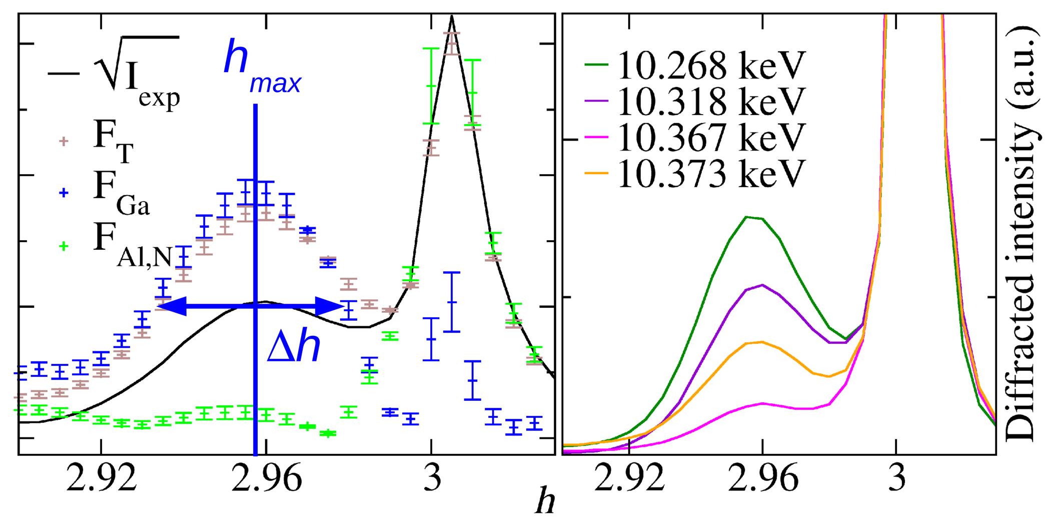


The incident angle of x-rays was set below the critical angle for total reflection in order to achieve **surface sensitivity** (upper 50-100 Å). Reciprocal space *h*-scans were measured around the *(30-30)* for a satisfactory sensitivity to **in-plane strains**. Two effects were studied : (i) the in-plane strain behaviour during the progressive capping of the QDs by AlN, and (ii) the vertical correlation effect regarding the QDs size as QDs layers are stacked.

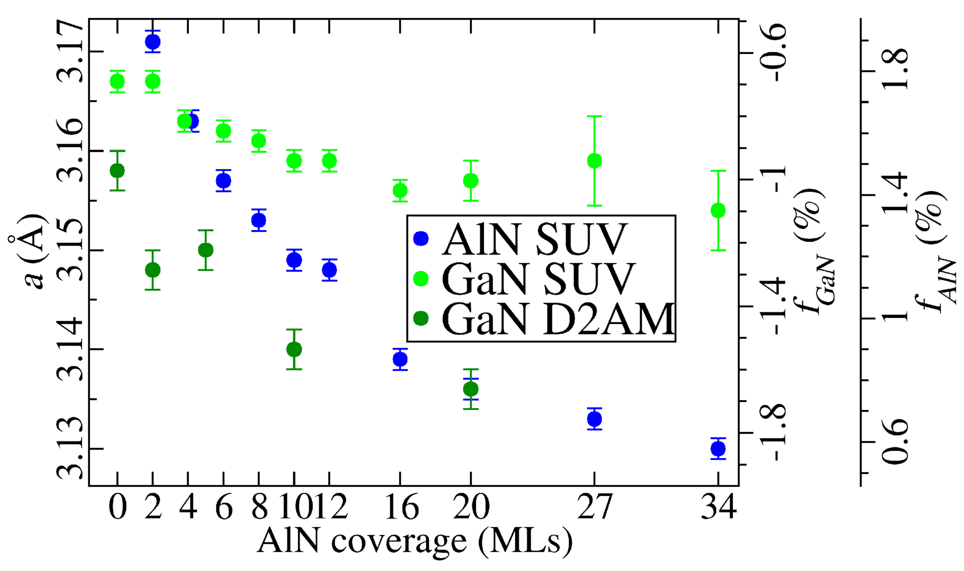
By means of **anomalous diffraction** at the Ga K-edge (10.367 keV) the contribution of the QDs to diffraction, as well as the one from AlN, can be distinguished. The diffracted intensity is measured at 11 energies around the Ga K-edge. The energy dependancy for every *h* value is fitted by[2]

$$I_{exp} \propto |F|^2 \propto |F_T|^2 + \frac{|F_{Ga}|^2}{f_{0,Ga}^2}\left(f'^2_{Ga} + f''^2_{Ga}\right) + 2\frac{|F_T||F_{Ga}|}{f_{0,Ga}}\left[f'_{Ga}\cos(\varphi_T - \varphi_{Ga}) + f''_{Ga}\sin(\varphi_T - \varphi_{Ga})\right]$$

F, φ : total | **F_T**, φ_T : non anomalous atoms + Thomson of Ga | **F_A**, φ_A : Thomson of Ga | **F_T &** φ_T-φ_A ⇒ **F_N** non anomalous (Al,N)

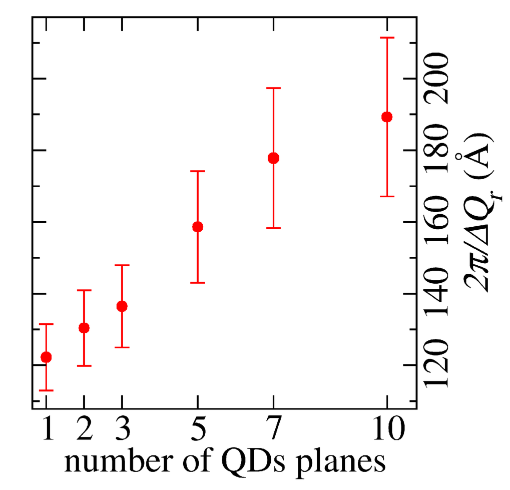


Diffracted intensities for 4 energies across Ga K-edge. F_{Ga} and $F_{Al,N}$ are separated from the total intensity. The average in-plane strain state is given by the position of the maximum (h_{max}) ; the average diameter is directly related to the width of F_{Ga} (Δh).



The experiments were carried out at the ESRF, *in situ* on BM32-SUV with 2 samples[3] (1 for capping effects, 1 for correlation effects), and *ex situ* on BM2-D2AM with a series of 5 samples with increasing AlN capping.

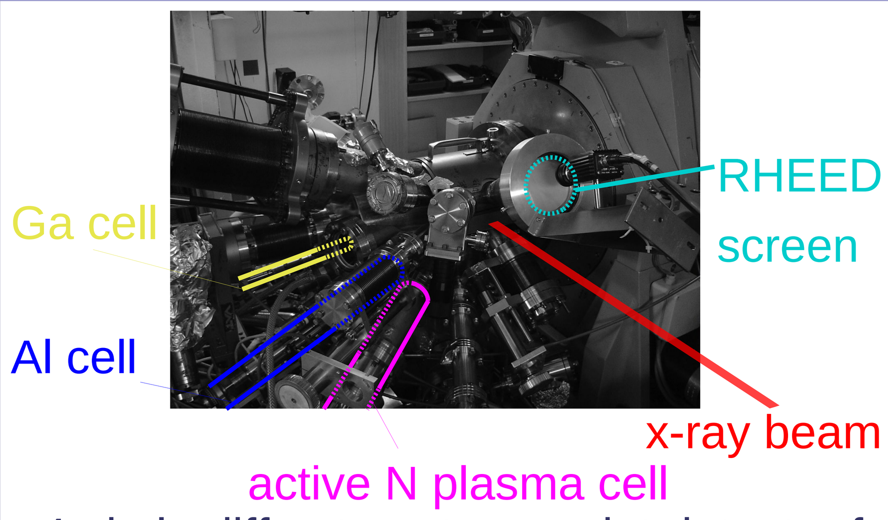
During the capping process, **QDs are progressively in-plane compressed as the AlN capping relaxes**.



A stagnation is observed above 30 MLs. This is possibly a limit above which vertical correlation effects in QDs layers stackings shall not happen.

A ~ 50 % average increase of the QDs diameter is demonstrated as a side effect of the vertical correlation in the position of QDs.

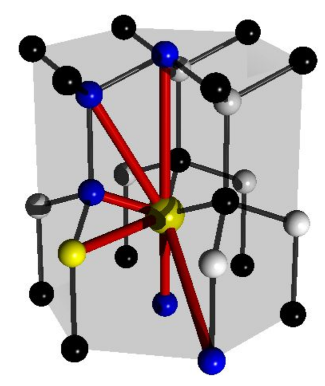
in situ growth setup



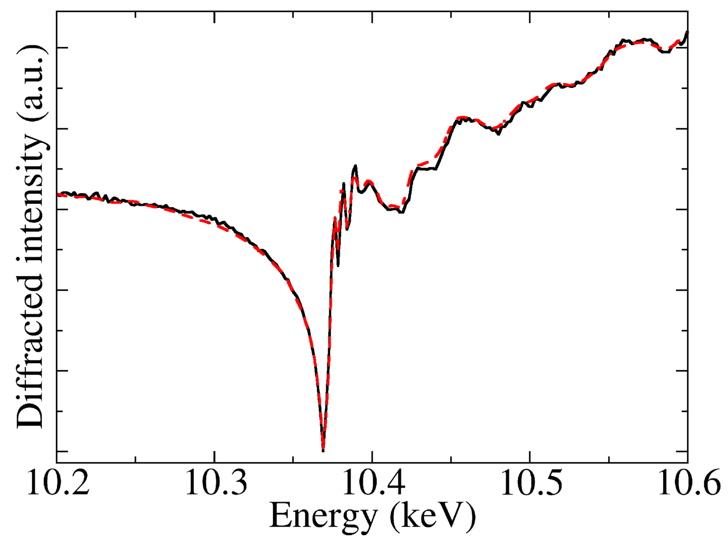
SUV (Surface Ultra Vacuum) apparatus on the BM32 beamline at ERSF dedicated to *in situ* growth and surface studies :

- **grazing incidence** setup
- RHEED, LEED, Auger
- 2D CCD camera for fast acquisition of **GISAXS** maps
- 4-circle diffractometer + point detector for **diffraction**
- energy tunab+++ility suitable for **anomalous** measurements

Diffraction Anomalous Fine Structure

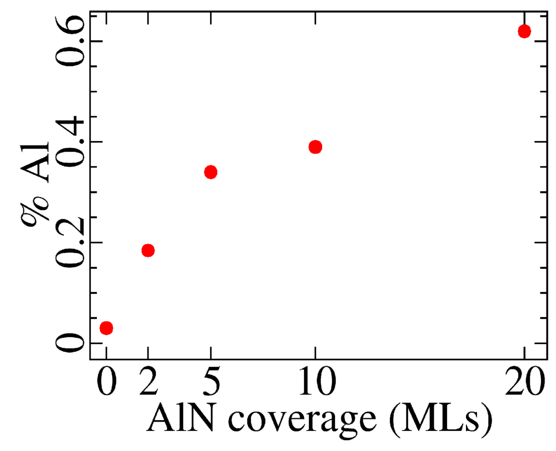


The **local environment of Ga atoms in the iso-strain regions** selected by diffraction was analysed by grazing incidence Diffraction Anomalous Fine Structure (DAFS) at the Ga K-edge. ***Q*** was fixed at *h=h$_{max}$*, that is, the average in-plane strain state in the QDs. Two aspects of the iso-strain region were investigated : (i) the out-of plane strain and (ii) the composition (Al, Ga, N).



Grazing incidence DAFS spectrum measured on the BM2-D2AM beamline at ESRF, and best crystallographic fit, for GaN QDs capped with 10 AlN MLs.

The edge profile was measured and fitted for a series QDs samples with increasing AlN capping. The only relevant fitting parameter was the Al composition. A linear increase up to 5 MLs suggests a **uniform capping process**, like a wetting of the QDs. The further evolution would indicate that **AlN fills up between the QDs**.

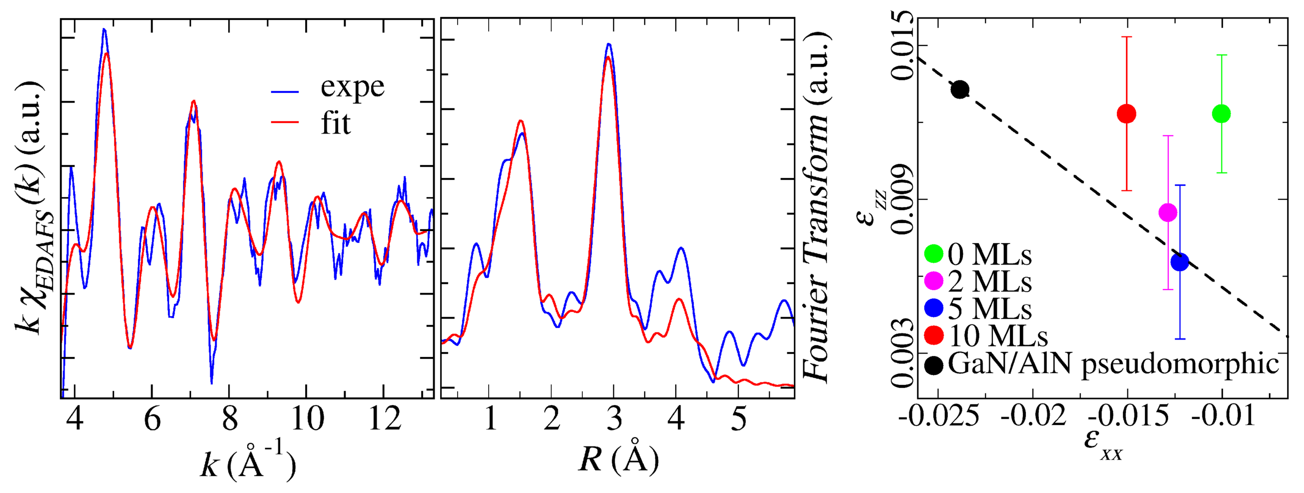


Extended DAFS oscillations were measured on BM2-D2AM and fitted using a EXAFS-like path formalism[4] :

$$\chi_{EDAFS} = \frac{\chi_Q}{S_D} \quad \text{and} \quad \chi_Q(k) = \sum_\gamma A_\gamma(k)\sin\left[2kR_\gamma + 2\delta_c(k) + \varphi - \varphi_{Ga} - \frac{\pi}{2}\right]$$

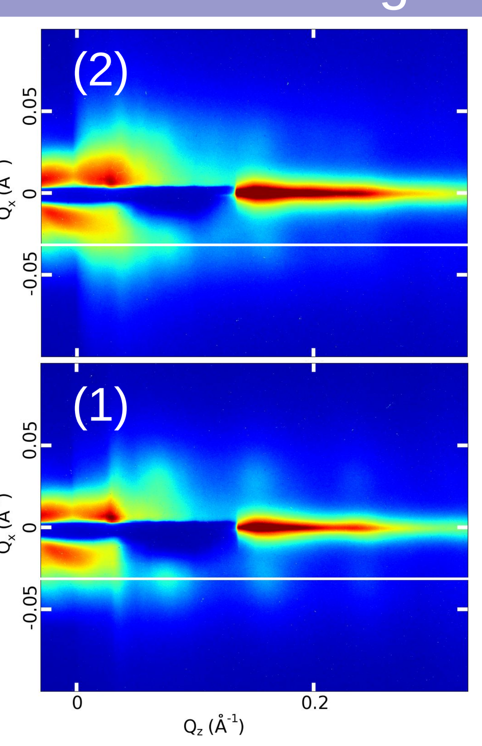
S_D : crystallo. normalization factor | γ : scattering paths | **A_γ** : net amplitude | δ_c : phase shift | **R_γ** : effective path length

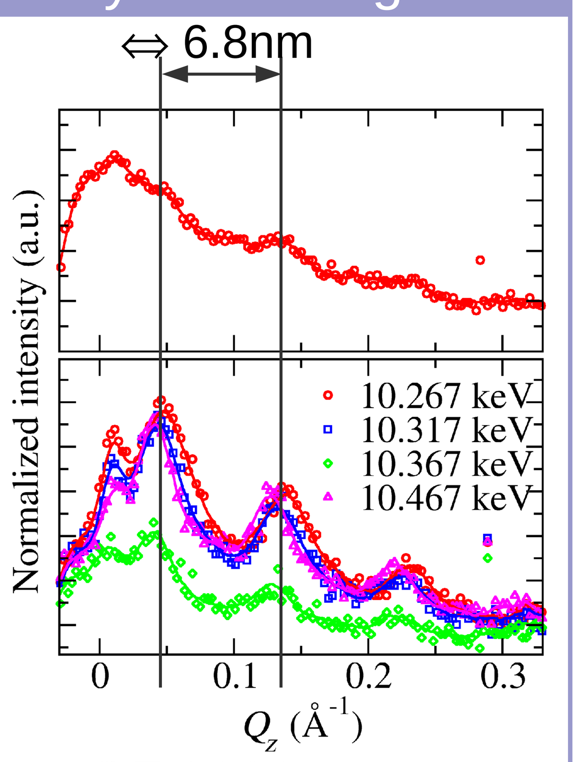
Five single scattering (red) and 2 multiple scattering paths were used to fit the experimental data in the [1.0 - 3.5] Å range, allowing the determination of the out-of-plane strain state as a function of the AlN capping thickness. A **discrepancy to the biaxial hypothesis** is observed.



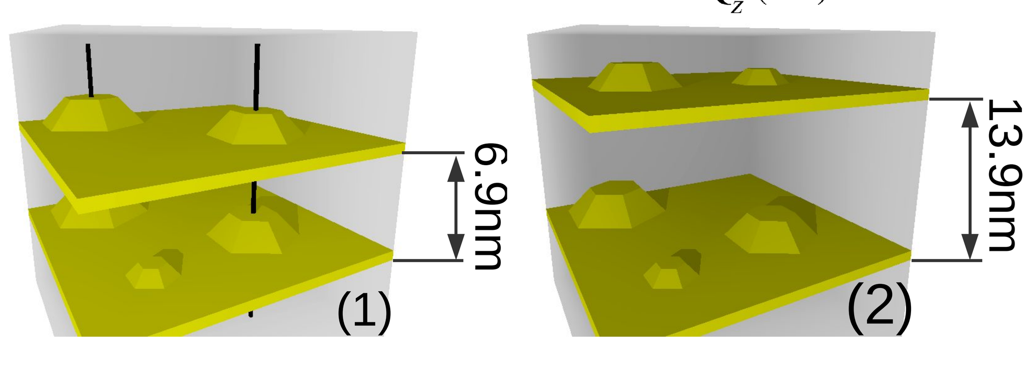
Anomalous Grazing Incidence Small Angle X-ray Scattering

Vertical correlation effects were demonstrated by ***in situ* Grazing Incidence Small Angle X-ray Scattering**[5] (GISAXS), which is sensitive to the **correlation lengths and morphology** of nanostructures. In particular, potential out-of-plane (vertical) correlation in the position of the QDs is expected along ***Q_z***.





GISAXS maps were measured on BM32-SUV for 2 stackings of QDs layers with distinct interlayer distances. The **critical incident angle** made the measurements sensitive to the whole stacking. GISAXS maps were measured at **several energies** around the Ga K-edge, to make sure the expected correlation effects originate from the QDs.



References

[1]N. Gogneau *et al.*, J. Appl. Phys. **94**, 2254 (2003)
[2]M.G. Proietti *et al.*, Phys. Rev. B **59**, 5479 (1999)
[3]J. Coraux *et al.*, submitted to Appl. Phys. Lett.
[4]M. Newville *et al.*, Phys. B **208-209**, 154 (1995)
[5]G. Renaud *et al.*, Nature **300**, 1416 (2003)