Step-by-step capping and strain state of GaN/AlN quantum dots studied by grazing-incidence
diffraction anomalous fine structure

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The investigation of small-size embedded nanostructures, by a combination of complementary anomalous
diffraction techniques, is reported. GaN quantum dots (QD’s), grown by molecular beam epitaxy in a modified
Stranski-Krastanow mode, are studied in terms of strain and local environment, as a function of the AlN cap
layer thickness, by means of grazing-incidence anomalous diffraction. That is, the x-ray photon energy is tuned
across the Ga absorption K edge which makes diffraction chemically selective. Measurement of hkl scans,
close to the AlN (3030) Bragg reflection, at several energies across the Ga K edge, allows the extraction of the
Ga partial structure factor, from which the in-plane strain of GaN QD’s is deduced. From the fixed-Q energy-
dependent diffracted intensity spectra, measured for diffraction-selected isostrain regions corresponding to the
average in-plane strain state of the QD’s, quantitative information regarding the composition and out-of-plane
strain has been obtained. We recover the in-plane and out-of-plane strains in the dots. The comparison to the
biaxial elastic strain in a pseudomorphic layer indicates a tendency to an overstrained regime.

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I. INTRODUCTION

Most of the interest in the improvement of the growth
techniques for III-V semiconductor nanostructures origin-
ates from the fact that the quantum confinement of carriers
leads to unique optoelectronic performances. The confinement in one dimension—that is, the growth of quantum wells
(QW’s)—has been brought under control for a variety of
systems, leading to QW’s based optoelectronic devices, such
as nitride QW laser diodes.1 However, due to the large den-
sities of defects in III-nitride materials,2 the emission effi-
ciency of such devices is strongly altered by increasing the
temperature. As an alternate way to overcome that difficulty,
carriers may be confined in regions free of any defect,3 such
as self-organized quantum dots (QD’s). For reasonable opto-
electronic efficiency, simultaneous control over the size, size
distribution, nucleation sites, density, and structure of the
QD’s is required. In the InAs/GaAs system, room-
temperature ultraviolet lasers4 could be achieved once these
requirements were satisfied. This article focuses on the
GaN/AlN system, for which the overall control of the QD
characteristics still remains a challenge. In the last few years,
molecular beam epitaxy (MBE) has recently been of particu-
lar interest with the improvement of QD density control, us-
ing the Stranski-Krastanow (SK) growth mode5–7 and
derivatives.8,10 Size homogenization of the QD’s was
achieved using vertical correlation through strain fields.11–13
Alternatively, new efforts are made to understand the ef-
facts of the QD capping by AlN, which strongly modifies the
strain state in the QD’s (Refs. 13 and 14) and therefore plays
a decisive role in modification of the optical properties.
Moreover, this topic raises a fundamental interest regarding
the physics of strain accommodation between a QD and its
capping.

Many complementary methods have been applied to
quantitative strain characterization in nanostructures.15 This
is true for grazing-incidence x-ray diffraction (GIXRD),
which can be made chemically sensitive when carried out as
a function of the energy across the absorption edge of an
element. This technique is known as anomalous diffraction16
and is suitable (a) to localize some element in reciprocal
space,17 (b) to identify the local environment of an atom,18
and (c) to determine the strain and composition of an iso-
strain region selected by diffraction.19

In this article we present a comprehensive strain analysis
of the capping of GaN QD’s by AlN. More precisely, we
study the in-plane and out-of-plane strain states and com-
position in the QD’s as a function of the AlN capping thick-
ness, by means of grazing-incidence anomalous x-ray dif-
fraction at the Ga K edge (10.367 keV), around the (3030)
reflection.

The MBE preparation of the set of samples to be analyzed
is detailed in Sec. II. The strain characterization technique,
by grazing-incidence diffraction anomalous fine structure
(GIDAFS), is detailed in Sec. III. The strain and composition
information obtained using different aspects of GIDAFS—
namely, multiwavelength anomalous diffraction (MAD), ex-
tended diffraction anomalous fine structure (EDAFS) oscil-
lations, and diffraction anomalous line-shape analysis are
given in Secs. IV–VI.

II. SAMPLES

The samples were grown in a MECA 2000 MBE chamber,
equipped with standard effusion cells providing the Ga and
Al fluxes and a radiofrequency plasma cell providing the
active nitrogen flux. The substrates were 2-μm-thick
AIN(0001) layers deposited by metal-organic chemical vapor deposition on sapphire.\textsuperscript{20} The substrate temperature was fixed at 740 °C. Prior to the growth of the QD’s, 10-nm-thick AlN buffers were grown. The QD growth was achieved in the modified SK growth mode,\textsuperscript{8,9} by depositing six GaN monolayers (ML) under Ga-rich conditions. This resulted in the formation of a Ga bilayer at the surface inhibiting the 2D-3D transition even above the usual \(~2\) ML critical thickness for the 2D-3D transition in the SK growth mode.\textsuperscript{5} The thermal evaporation under vacuum of the Ga bilayers led to the transition of the 2D GaN layer into 3D QD’s (Ref. 8 and 9) connected by a \(~2\)-ML-thick wetting layer (WL). A set of five samples was grown with increasing AlN capping: 0, 2, 5, 10, and 20 ML. Figure 1 shows a \(1-\mu m^2\) AFM image of free-standing dot samples. The height of the QD’s was evaluated to \(3.0\pm0.5\) nm and their diameter to \(15\pm1\) nm, leading to an aspect ratio of about 0.2. The QD density was found as high as \(1.3\times10^{11}\) cm\(^{-2}\); that is, QD’s are almost adjacent. The roughness of the samples was measured to follow the evolution of the morphology as a function of the AlN deposit. Figure 2 shows the evolution of the root-mean-square (rms) roughness for 0, 2, 5, and 10AlN ML deposited. The rms roughness remains roughly the same until a 5-ML deposit and drops for the 10-ML sample.

**FIG. 1.** \(1-\mu m^2\) AFM images of uncapped GaN QD’s grown in the modified SK mode.

**FIG. 2.** (Color online) rms roughness evolution as a function of the AlN deposit amount.

Grazing-incidence diffraction anomalous fine structure at the Ga \(K\) edge (10.367 keV) was performed at the French Collaborative Research Group beamline BM2 at the European Synchrotron Radiation Facility (ESRF) by using the eight-circle diffractometer equipment. We measured the diffuse scattering intensity, in grazing incidence and exit, close to the in-plane (30\(\overline{3}\)0) Bragg reflection of the AlN substrate (radial scans), at energies close to the Ga \(K\) edge. Figure 3 sketches the experimental setup. The samples were mounted in the vertical plane; i.e., the polarization vector \(i\hat{\vec{e}}\) of the incident photon beam was perpendicular to the sample surface (0001). The incidence angle was \(\alpha_i=0.17^\circ\), lower than the bulk AlN critical angle \(\alpha_c=0.21^\circ\) (at 10.32 keV) for which the total reflection regime takes place. Such conditions were used to enhance the weak contribution of the encapsulated dot layer with respect to that of the substrate. The diffraction geometry was chosen in such a way as to keep the scattering vector in the vertical plane. The diffraction point detector was a scintillator, and the slits were opened so as to measure the integrated intensity over the grazing exit angle \(\alpha_f\). A photodiode measuring the fluorescence yield of an in-vacuum 4-\(\mu\)m Ti foil was used to monitor the incoming x-ray beam. Two kinds of scans were performed: (a) \(h\) scans (radial) in the range 2.9–3.05 for 12 energies, from 10.272 to 10.472 keV—i.e., close to the Ga \(K\) edge—and (b) energy scans at fixed scattering vector (Q) corresponding to the maximum of the QD contribution to the diffuse scattering (i.e., at the maximum of the partial structure factor \(F_{\text{AsGa}}\) profile extracted from the multilayer profile extracted from the multilayer \(h\) scans; see the following section and Ref. 19). The energy scans were recorded in a large energy interval, typically 1 keV, with an energy step from 1 to 2 eV, to allow a quantitative analysis of both the edge and extended oscillations.

**III. GIDAFS MEASUREMENTS**

The solid lines in Figs. 4(a), 4(c), and 4(e) show the square-root intensities along the [10\(\overline{1}\)0] direction \((h\) scan) close to the (30\(\overline{3}\)0) reflection as a function of the AlN coverage. These \(h\) scans are related to both the in-plane strain state and size. With no AlN coverage, one observes a diffuse-scattering peak ascribed to QD’s slightly strained by the AlN...
buffer and substrate. As the AlN coverage increases [from (a) free-standing QD’s to (e) 10-ML AlN coverage], this peak is progressively shifted towards higher h values and gets mixed with the AlN buffer peak. Further analysis was made possible by distinguishing the GaN and AlN contributions, using MAD measurements.\textsuperscript{16,17,19} Figures 4(b), 4(d), and 4(f) show some of the square-root diffracted intensities measured for increasing AlN coverage, across the Ga K edge, taking advantage of the Ga anomalous effect to localize the Ga contribution along [10\overline{1}0]. The Ga scattering factor can be written as $f_{\text{Ga}}=f_{\text{Ga}}^0+f_{\text{Ga}}^\alpha+i\beta_{\text{Ga}}$, where $f_{\text{Ga}}^0$ and $f_{\text{Ga}}^\alpha$ are the Ga real and imaginary anomalous (resonant) scattering corrections and $f_{\text{Ga}}^0$ is the Ga Thomson scattering factor. From MAD measurements, the Ga partial structure factor $F_{\text{Ga}}$ of phase $\varphi_{\text{Ga}}$, which includes the Thomson scattering of all anomalous atoms (Ga), can be retrieved. The retrieval shall be run in the framework of the distorted-wave Born approximation, taking into account scattering paths involving the reflection from the layer supporting the dots.\textsuperscript{21,22} We recorded the diffracted intensity integrated over the exit angle $\alpha$, between 0 and 2$\alpha$, and therefore collected all the scattering paths. Discarding the energy dependence of the reflection coefficients at the Ga K edge, as a consequence of the small Ga amount (six equivalent monolayers), the recorded intensity corrected for fluorescence, $I_{\text{exp}}$, is proportional to the total square structure factor $||F||^2$:

$$I_{\text{exp}}(E) \propto ||F||^2 \approx ||F_{\text{Ga}}||^2 \left[ \cos(\varphi_T-\varphi_{\text{Ga}}) + \beta_{\text{Ga}}' ||F_{\text{Ga}}||^2 \right]$$

$$+ \left[ \sin(\varphi_T-\varphi_{\text{Ga}}) + \beta_{\text{Ga}}' ||F_{\text{Ga}}||^2 \right],$$

where $\beta=||F_{\text{Ga}}||/||F_{\text{Ga}}||/||F_{\text{T}}||$. Figure 5 shows the total and partial structure factor relations in the complex plane. The partial structure factor $F_{\text{T}}$ of phase $\varphi_{\text{T}}$ that includes the overall contribution of nonanomalous atoms and the Thomson scattering of all anomalous atoms, $F_{\text{Ga}}$, as well as $\varphi_T-\varphi_{\text{Ga}}$, can be extracted for all h values, without any structural model by fitting Eq. (1) to the experimental data with the NANOMAD algorithm.\textsuperscript{23}

As shown in Figs. 4(a), 4(c), and 4(e), $F_{\text{Ga}}$ and $F_{\text{T}}$ were extracted. The h=0 position of the diffuse $F_{\text{Ga}}$ peak maximum is inversely proportional to the in-plane average lattice parameter $a_{\text{GaN}}$, since the distance between GaN (30\overline{3}0) planes is $d_{30\overline{3}0}=3(\sqrt{3}/2)a_{\text{GaN}}/3=3(\sqrt{3}/2)a_{\text{AlN}}/h_{\text{Ga}}$ with $a_{\text{AlN}}=3.112$ Å, for the AlN substrate peak used as a reference. Figure 6 shows the evolution of the in-plane lattice parameter as a function of the AlN cap thickness. The uncapped QD’s are partially in-plane relaxed, with an average strain relative to bulk GaN, $\varepsilon_{\text{GIXRD}}=(a_{\text{GaN,GIXRD}}-a_{\text{GaN,bulk}})/a_{\text{GaN,bulk}}\approx-1\%$, with $a_{\text{GaN,bulk}}=3.189$ Å. The QD’s are then progressively in-plane compressed by the AlN capping, but remain slightly relaxed, $\varepsilon_{\text{GIXRD}}\approx-1.6\%$, compared to pseudomorphic GaN ($\varepsilon_{\text{GIXRD}}\approx-2.4\%$).

V. EDAFS ANALYSIS

Out-of-plane information can be achieved by a quantitative analysis of the grazing-incidence diffraction anomalous fine structure oscillations in the extended region above the edge (EDAFS). Figure 7(a) shows for free-standing QD’s the oscillatory contribution ($\chi_{\text{EDAfs}}$) to the DAFS spectrum, extracted and normalized to the smooth atomic background ($I_0$):

\begin{figure}[h]
\centering
\includegraphics[width=\linewidth]{fig5.pdf}
\caption{Schematic representation in the complex plane of the structure factor F as a function of $F_{\text{T}}$, $F_{\text{A}=\text{Ga}}$, and $\varphi_T-\varphi_{\text{A}=\text{Ga}}$ (see text). $F_N$ represents the partial structure factor of nonresonant atoms (Al, N).}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\linewidth]{fig6.pdf}
\caption{In-plane lattice parameter $a_{\text{GaN,GIXRD}}$ and strain (relative to bulk GaN) in GaN deduced from the position of the $F_{\text{Ga}}$ maximum. Bulk GaN gives $\varepsilon_{\text{GIXRD}}=0\%$ with $a_{\text{GaN,bulk}}=3.189$ Å while bulk AlN gives $\varepsilon_{\text{GIXRD}}=-2.4\%$ with $a_{\text{AlN}}=3.112$ Å.}
\end{figure}
\[ \chi_{\text{DAFS}} = \frac{I_{\text{exp}} - I_0}{I_0}. \]

\( \chi_{\text{DAFS}} \) can be written as \( \chi_{\text{DAFS}} = \frac{1}{2} \chi_Q \) where \( S_0 \) is a normalization factor that depends on crystallography and is calculated from the parameters \( \Delta \varphi = \varphi_j - \varphi_A \) and \( \beta \) [see Eq. (1)] and \( \chi_Q \) is in the first-order approximation of the diffracted anomalous fine structure:

\[ \chi_Q(k) = \cos(\varphi_0 - \varphi_A) \sum_{j=1}^{N_1} w_j' \chi_j' + \sin(\varphi_0 - \varphi_A) \sum_{j=1}^{N_2} w_j'' \chi_j'', \]

where \( \varphi_0 \) is the phase of the smooth structure factor (without oscillations), the \( j \) label runs over the different anomalous sites \( \Lambda \) (the Ga sites, \( \Lambda = \text{Ga} \)) and \( w_j' = \frac{\{P_j\} \cos(\varphi_j - \varphi_A)}{\sin(\varphi_j - \varphi_A)} \) and \( w_j'' = \frac{\{P_j\} \sin(\varphi_j - \varphi_A)}{\sin(\varphi_j - \varphi_A)} \) are crystallographic weights. The term \( \chi_j' (\chi_j'') \) in Eq. (2) is the oscillatory part of the resonant atomic scattering factor \( f_j' (f_j'') \); it is related to the local atomic environment of the resonant atom. \( \chi_j'' \) is formally identical to the extended x-ray absorption fine structure (EXAFS) oscillations of atom \( j \). In the present case—i.e., one statistically equivalent site—\( \chi_Q \) can be rewritten as a function of the virtual photoelectron wave-vector modulus \( k \) in a form that is similar to the well-known EXAFS formula:

\[ \chi_Q(k) = \sum_{\gamma} A_{\gamma}(k) \sin \left[ 2k(R) + \varphi_{\gamma}(k) + 2 \delta_{\gamma}(k) + \varphi_0 - \varphi_A - \frac{\pi}{2} \right]. \]

where \( \gamma \) runs over all possible virtual photoelectron scattering paths, \( \langle R \rangle \), is the effective length of path \( \gamma \) and \( \varphi_{\gamma}(k) \) is the net scattering photoelectron phase shift.

The analysis can be performed according to the standard criteria and available codes for EXAFS, provided that crystallographic phases and amplitude correction factors are taken into account (for more details see Refs. 24 and 25). The EDAFS analysis has been carried out by using the FEFF8 code \(^{26}\) to generate theoretical phases and amplitudes, taking into account beam polarization, for a 6-Å-radius GaN cluster. In order to address the possible presence of Al atoms in the QD’s or at the substrate and capping interface, Ga-Al and Ga-N-Al scattering paths were considered by calculating an AlN cluster with the Ga central atom as absorber. The ARTEMIS interface to the IFEFFIT package \(^{27}\) was used to fit theoretical computations to the experimental data.

The EDAFS spectra were Fourier transformed in the \( k \) range 3–10 Å\(^{-1}\), and the fit was performed in \( R \) space (real space), using four next-neighboring shells (I–IV). As an example, we show the best-fit curves for free-standing QD’s in Figs. 7(a) and 7(b), compared to the experimental raw data. Six single scattering (SS) paths and four multiple-scattering (MS) paths were found to be relevant in this range (see Fig. 8).

(i) \((\text{Ga-N})_1\), in-plane, I-shell path, corresponding to the three Ga-N bonds of the tetrahedron that are nearly in plane.

(ii) \((\text{Ga-N})_1\), out-of-plane, I-shell path, corresponding to the fourth Ga-N bond of the tetrahedron, lying along the \( c \) axis.

(iii) \((\text{Ga-Ga})_2\), II-shell, out-of-plane path, corresponding to six Ga atoms at a distance that is a combination of \( a \) and \( c \), \( \{ \frac{1}{2} a_{\text{GIXRD}}^2 + \frac{1}{4} c^2 \}^{1/2} \), where \( a_{\text{GIXRD}} \) is the in-plane lattice pa-

FIG. 7. (a) Experimental EDAFS for the free-standing QD sample, compared with the best-fit result and (b) \( R \)-space experimental curve for free-standing QD’s compared with the best fit.

FIG. 8. Scheme of GaN wurzite structure; the most relevant virtual photoelectron scattering paths used for the EDAFS simulation are represented: (1) in-plane I-shell \( (\text{Ga-N})_1 \), (2) out-of-plane I-shell \((\text{Ga-N})_1\), (3) out-of-plane II-shell \((\text{Ga-Ga})_2\), (4) III shell Ga-Ga along \( c \), (5) nearly in-plane IV-shell Ga-N, MS Ga-N-N, and Ga-N-Ga. Ga atoms are represented by white spheres, N by black ones.
TABLE I. EDAFS best-fit values for interatomic distances ($R$), Debye-Waller factors ($\sigma$), and Al concentration ($x_{Al}$) obtained by IFFIT minimization using theoretical fitting standards provided by the FEFF8 code. The amplitude and phase correction factors have been obtained by crystallographic analysis of the DAFS line shape. The $a_{GaN,GIXRD}$ value is kept fixed to the value determined by grazing incidence and exit diffraction (diff.).

<table>
<thead>
<tr>
<th></th>
<th>Bulk</th>
<th>GaN/AlN</th>
<th>0 MLs</th>
<th>2 MLs</th>
<th>5 MLs</th>
<th>10 MLs</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R_1$(Ga-N) (Å)</td>
<td>-</td>
<td>-</td>
<td>1.93</td>
<td>1.94</td>
<td>1.94</td>
<td>194</td>
</tr>
<tr>
<td>$\sigma_1^2$(Å$^2$)</td>
<td>-</td>
<td>-</td>
<td>$2 \times 10^{-3}$</td>
<td>$4 \times 10^{-3}$</td>
<td>$4 \times 10^{-3}$</td>
<td>$1 \times 10^{-3}$</td>
</tr>
<tr>
<td>$R_2$(Ga-Ga)=$a_{GaN}$ (Å)</td>
<td>3.188</td>
<td>3.11</td>
<td>3.156 (diff.)</td>
<td>3.147 (diff.)</td>
<td>3.149 (diff.)</td>
<td>3.14 (diff.)</td>
</tr>
<tr>
<td>$\sigma_2^2$(Å$^2$)</td>
<td>-</td>
<td>-</td>
<td>$6 \times 10^{-3}$</td>
<td>$8 \times 10^{-3}$</td>
<td>$4 \times 10^{-3}$</td>
<td>$7 \times 10^{-3}$</td>
</tr>
<tr>
<td>$c_{GaN}$ (Å)</td>
<td>5.186</td>
<td>5.26</td>
<td>5.25±0.02</td>
<td>5.23±0.03</td>
<td>5.22±0.02</td>
<td>5.25±0.04</td>
</tr>
<tr>
<td>$c_{GaN}/a_{GaN}$</td>
<td>1.626</td>
<td>1.69</td>
<td>1.66</td>
<td>1.66</td>
<td>1.66</td>
<td>1.67</td>
</tr>
<tr>
<td>$x_{Al}$</td>
<td>-</td>
<td>-</td>
<td>0.1±0.1</td>
<td>0.0±0.1</td>
<td>0.1±0.1</td>
<td>0.05±0.1</td>
</tr>
</tbody>
</table>

parameter obtained with the grazing-incidence (and exit) x-ray diffraction experiment (see Sec. IV).

(iv) Ga-N, III-shell path, corresponding to one N atom along the c direction.

(v) Ga-N, IV-shell path, corresponding to six N nearly in-plane atoms.

(vi) MS paths consisting of triangular paths Ga-N-N and Ga-N-Ga.

The “in-plane” statement refers to the surface or growth plane, and all the scattering paths, except the first one, were expressed in terms of $a$ and $c$ cell parameters, as requested by the hexagonal cell symmetry.

We performed the fit by fixing the $a$ parameter (in plane) to the values found by diffraction, $a_{GaN,GIXRD}$, letting the $c$ parameter vary according to the hexagonal symmetry. The Ga-N first-shell distances were let free to vary independently of $a$ and $c$ since, as is well known, Vegard’s law is far from being valid for semiconductor alloys, in which the bond-bending mechanism is dominant compared to bond stretching.28 The presence of Al is taken into account by adding the correspondent scattering paths in which Al substitutes for Ga as next nearest neighbor (NNN) and multiplying the amplitude by a factor $x_{Al}$ for Al and $(1-x_{Al})$ for Ga. The best-fit parameters are shown for the whole set of samples in Table I, where we also report, as a reference, the bulk and pseudomorphic values for GaN.29 The Ga-Al distance was also let free to vary and the values found were close to the Al-Al NNN distance. Since the Al content is found to be zero within the statistical errors, the Al-Al NNN distance was not reported in the table. Starting from the fit results, the in-plane and out-of-plane strains were calculated, with respect to relaxed (bulk) GaN, as $e_{xx,GIXRD}=(a_{GaN,GIXRD}-a_{GaN,bulk})/a_{GaN,bulk}$ and $e_{zz,GIDA}=c_{GaN,GIDA}-c_{GaN,bulk})/c_{GaN,bulk}$. $e_{xx,GIXRD}$ vs $e_{zz,GIDA}$ for the different samples studied is sketched in Fig. 9. These values are compared to the biaxial elastic behavior for pseudomorphic GaN on AlN (straight line), which corresponds to $e_{xx}=-2e_{c_1}c_{13}/c_{13}$, with the elastic coefficients $c_{13}$ and $c_{13}$ values from Ref. 30.

We observe the following general findings.

(i) The Ga-N first-shell in-plane and out-of-plane distances are seen to be very close to each other, within the fit errors (0.01 Å), in agreement with previous studies.31

(ii) As shown in Table I, the Al content remains very small, showing that no intermixing takes place in the QD’s as expected for the Al/Ga species.32

(iii) The $c_{GaN,GIDA}$ values range from 5.22 to 5.25 Å, which is quite large compared to the values foreseen by the elastic regime of a pseudomorphic GaN layer, as apparent in Fig. 9 where the experimental points fall above the elasticity curve.

As expected, the uncapped QD’s do not follow a biaxial strain behavior, because of the presence of a free surface. However, capping by a thin layer (2–5 ML) of AlN should favor evolution towards the biaxial case for at least two reasons: (a) first of all, capping results in a size decrease of the dots—i.e., an aspect ratio reduction associated with a relative increase of the biaxial component of the strain33—(b) furthermore, we speculate that the possible wetting of the QD’s by AlN, which will be discussed in Sec. VI, strongly tends to reduce the relaxation through the free surface, which also results in a relative increase of the biaxial component of the strain. Besides these considerations, it is worth noting that the plastic relaxation process of AlN deposited on GaN, which is characterized by a very small critical thickness,34 is still unclear and may also determine to some extent the strain state of the AlN/GaN QD interacting system.

FIG. 9. GaN QD strain $e_{xx}=(a_{GaN,GIXRD}-a_{GaN,bulk})/a_{GaN,bulk}$ vs $e_{zz}=(c_{GaN,GIDA}-c_{GaN,bulk})/c_{GaN,bulk}$ values for all the samples studied compared with elastic biaxial strain of a pseudomorphic GaN thin film.
VI. EDGE ANALYSIS

The diffraction anomalous spectra, close to the Ga K edge, can give the Al and Ga relative compositions inside the GaN/AlN in-plane isostrain region selected with grazing incidence and exit diffraction; this region includes the GaN QD’s and the AlN on top. Previous studies clearly indicate that no atomic intermixing occurs at the GaN/AlN interfaces, with neither GaN/AlN QW’s nor GaN/AlN QD’s. This is confirmed by the grazing-incidence EDAFS results reported in Sec. V, which clearly show no significant Al/Ga mixing.

On the other side, analysis of the DAFS edge shape can give information about the capping mechanism of the QD’s. Indeed, as a first approximation, the diffused intensity is proportional to the square modulus of the in-plane isostrain region structure factor. We calculate this structure factor for an Al$_x$Ga$_{1-x}$N wurtzite structure to take into account the Al atoms belonging to the same isostrain region as the Ga atoms at the QD top. The Al concentration obtained by refining the $x$ value given the Al atom fraction seen by diffraction, contributing at the chosen $Q$ value and determining the edge line shape. Taking into account that EDAFS analysis shows that no intermixing takes place, we can state that we are probing the AlN capping. Figure 10(a) shows the GIDAFS spectra for the 0, 2, 5, and 10 ML AlN cap thicknesses, measured at the maximum of the partial structure factor $F_{x=0.39}$ (see Sec. IV). The data were normalized so that the intensity at 10.2 keV is the same for all spectra. Equation (1) was fitted to each GIDAFS spectrum, using the anomalous scattering factors $f'_{Ga}$ and $f'_{Al}$ of a GaN layer. A scale factor, the detector efficiency as a function of the energy and the Al occupation factor ($x$) inside the in-plane isostrain region, was refined. As an example, Fig. 10(b) shows the best fit for the 10-ML sample obtained with $x=0.39\pm0.01$. It should be noted that the occupation factor is determined by the ratio $\beta$

![FIG. 10. (Color online) (a) GIDAFS spectra for 0-, 2-, 5-, and 10-ML AlN capping, measured at maximum of $F_{x=0.39}$ and (b) crystallographic best fit for the 10 ML AlN sample. Open circle: experiment. Solid line: simulation performed with experimental $f'_{Ga}$ and $f'_{Al}$ of a GaN thin film.](image)

![FIG. 11. (Color online) Al-atoms occupation factor $x$ of the Al$_{x}$Ga$_{1-x}$N isostrain region selected by diffraction (maximum of $F_{Ga}$), as a function of AlN coverage. Inset: evolution of the AlN thickness on top of the dots plotted as a function of the nominal AlN deposit (see text). The dashed straight line of slope 1 corresponds to an AlN quantity on dots equal to the nominal AlN deposit.](image)
VII. CONCLUSION

We have presented results on the structural properties of GaN QD’s by combining different aspects of x-ray diffraction: quasi surface sensitivity due to grazing incidence, quantitative analysis of anomalous effects according to MAD principles, line-shape fit of DAFS and EDAFS oscillations fit.

All these aspects are strongly complementary. We determine in-plane and out-of-plane lattice parameters and investigate the effect of the capping layer by monitoring its effect on the QD strain. In addition, the Al fraction seen by the anomalous diffraction as a function of the capping layer thickness (obtained by GIDAFS line-shape analysis at the Ga K edge) indicates a wetting of the QD’s, followed by a noticeable change in the capping process which may be related either to plastic relaxation in AlN, to spatially selective AlN growth, or to a still unknown process. Let us point out that the Al fraction obtained in this way does not represent the Al content inside the dots, but the AlN contribution to the diffuse scattering at the same Q value as the GaN QD contribution—i.e., AlN mostly located on top of the QD’s. The Al content of the dots can be found by analysis of the EDAFS oscillations which provide the microscopic local environment of the Ga resonant atom. Our analysis shows that no Ga/Al intermixing takes place, as expected for these two group-III-N elements. We recovered the in-plane and perpendicular strains $e_{xx}$ and $e_{zz}$ in the dots and compare them to the biaxial elastic strain of a pseudomorphic layer. We find a tendency to an overstrained regime that suggests a more complex mechanism of strain accommodation which deserves further investigations.

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