Implantation damage formation in a-, c- and m-plane GaN

K. Lorenz a,⁎, E. Wendler b, A. Redondo-Cubero a, c, N. Catarino a, M.-P. Chauvat d, S. Schweiger e, F. Scholz e, E. Alves a, P. Ruterana d

a IPN, Instituto Superior Técnico, Universidade de Lisboa, Campus Tecnológico e Nuclear, Estrada Nacional 10, 2695-066, Bobadela LRS, Portugal
b Friedrich-Schiller-Universität Jena, Institut für Festkörperphysik, Max-Wien-Platz 1, 07743, Jena, Germany
c Departamento de Física Aplicada y Centro de Micro-Análisis de Materiales, Universidad Autónoma de Madrid, Madrid, Spain
d Centre de recherche sur les Ions les Matériaux et la Photonique (CIMAP) ENSICAEN, Boulevard Maréchal Juin, 14050, Caen, France
e Institute of Optoelectronics, University of Ulm, Albert-Einstein-Allee 45, D-89069, Ulm, Germany

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Abstract

Epitaxial GaN layers with a-, c- and m-plane surface orientations were implanted with 300 keV Ar-ions at 15 K with fluences ranging from 2 × 1012 to 4 × 1016 at/cm². Damage build-up proceeds in three steps separated by wide fluence regions where the maximum damage level, measured by in situ Rutherford Backscattering Spectrometry/Channelling, saturates. The three steps occur at similar fluences for the three crystal orientations and similar defect formation rates for the lowest fluences are observed. Surprisingly, the second saturation regime reveals a significantly lower damage level in a-plane layers while m- and c-plane samples suffer more than 4 times higher damage levels. The strong radiation resistance of a-plane GaN was attributed to very efficient dynamic annealing of point defects during the implantation even at 15 K. The migration and aggregation of point defects also lead to distinct defect microstructures as evidenced by transmission electron microscopy. Besides point defects and their clusters the dominant extended defects caused by implantation in c-plane GaN are basal stacking faults while dislocation loops are formed in a-plane material. m-plane GaN presents a mixture of planar defects and dislocation loops after implantation.

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1. Introduction

Despite the overwhelming success of blue light emitting diodes (LEDs) and lasers based on InGaN/GaN, III-nitride devices emitting at longer wavelength still suffer from low efficiencies, in part, due to strong polarization effects in conventional c-plane wurtzite materials [1,2]. Non-polar GaN, grown in the a- or m-plane of the wurtzite lattice, are known to avoid these polarization effects [3]. In addition, non-polar devices open the way for new applications due to polarized light emission [4]. Recently, ion implantation was successfully employed in the fabrication of non-polar vertical-cavity surface-emitting lasers where Al-implantation into m-plane GaN was used to define an aperture for lateral confinement allowing a significant decrease of the lasing threshold current density [5]. In c-plane nitrides, implantation was successfully used for doping and implant isolation of transistor structures as well as for the definition of mask-less areas for selective growth [6–8].

Despite these promising results, implantation damage build-up processes in III-nitride semiconductors are still poorly understood, in particular for materials other than c-plane GaN such as ternary alloys or thin films grown along other crystal directions [9]. In fact, some applications, such as doping, require high implantation fluences for which implantation damage becomes an important limitation. The damage accumulation processes in c-plane GaN have been investigated by many groups and were reviewed for example in refs. [9–13]. The main characteristics can be summarized as followed: In a wide range of implantation conditions (ions, fluences, temperatures etc.), implantation causes a bimodal damage distribution with a pronounced defect accumulation at the surface and another defect peak at a deeper range in the crystal bulk [10]. The surface damage was attributed to the fact that the surface acts as a sink for migrating point defects [10]; for high fluences, this typically leads to the formation of a highly damaged layer consisting of randomly oriented nanocrystals, possibly admixed with voids and amorphous regions [14]. For room temperature implantation, the bulk damage typically saturates at high fluences and no amorphous state is reached. Only for few ion...
species a complete disruption of the single crystal is observed due to
to chemical effects (e.g. O and C [11]) or for extremely high fluences
but low sputter rates [13]. For heavy ions and high fluences, sput-
tering of the nanocrystalline surface layer takes place and signifi-
cantly affects the defect profiles [15]. Strong dynamic annealing
processes lead to damage build-up curves with fluence exhibiting
several steps and wide fluence regions where defect levels saturate
even at cryogenic temperatures as low as 15 K [16]. Besides defect
annealing, the high mobility of point defects promotes, on the other
hand, the formation of extended defects such as defect clusters,
dislocation loops and stacking faults [17,18,13]. Implantation de-
fects were shown to introduce large hydrostatic strains in GaN and
related alloys [19–23]. Recently the interest in measuring this
strain has strongly increased since it was suggested to be one of the
driving forces for the transformation between distinct defect mi-
crostructures in different fluence regimes [24]. This assumption is
in particular backed by the fact that very similar damage formation
mechanisms act at 15 K and at room temperature [13,25]. However,
other effects, such as the deposition of energy due to electronic
energy loss, also need to be taken into account.

Recently, Catarino et al. suggested that a-plane GaN is more
resistant to implantation damage than c-plane GaN [26]. In this
report, implantation damage was studied using Rutherford back-
scattering Spectrometry/Channelling (RBS/C) which numbers the
number of displaced atoms perpendicular to the channelling axis,
the a-direction in this case. However, by measuring RBS/C only, it is
difficult to decide if indeed the nature and density of implantation
defects changes or if the same defect configuration leads to
different backscattering yields along the various crystal axes due to
shadowing by the matrix atoms. The understanding of implanta-
tion damage formation for non-polar surfaces may also become
important to understand ion implantation processes in nanowires
where the non-polar facets form the majority of the surface [27].

In the present work we compare the implantation damage build-up
at cryogenic temperature in GaN films grown along three
different crystallographic directions, namely, a-plane, c-plane
and m-plane GaN (in the following called a-, c- and m-GaN).
Comparison of in situ RBS/C at 15 K and ex situ transmission electron mi-
croscopy (TEM) results suggests that, despite similar damage
accumulation rates for low fluences, at high fluences, the nature of
extended defects changes leading to significant differences in the
defect profiles for samples with different surface orientation.

2. Experimental details

Nominally undoped c- and a-plane GaN films were grown by
Metal Organic Vapour Phase Epitaxy (MOVPE). c-GaN, ~2 μm thick
and grown on c-sapphire, was purchased from CREE. a-GaN was
grown on r-sapphire using a 20 nm AlN nucleation layer followed by
5.2 μm a-oriented GaN. The growth of a-GaN was interrupted
twice, after 300 and 1000 nm, to deposit a SiN intermediate layer
[28]. m-GaN, ~10 μm thick, was grown by Hydride Vapour Phase
Epitaxy (HVPE) on a m-GaN/m-SiC pseudo-template [29].

Implantation and in situ RBS/C analysis were carried out at the
Jena double beam chamber which allows sequential implantation
steps and consecutive RBS/C analysis without changing the sample
temperature [30]. The implantation was performed at 15 K using
300 keV argon (Ar) ions impinging at an angle 7° off the surface
normal in order to suppress channelling effects. Fluences ranged
from 2 × 1012 to 4 × 1016 at/cm² leading to a maximum Ar con-
centration of 2.7 at% for the highest fluence as estimated with
Monter Carlo simulations using the code SRIM [31]. The low im-
plantation and measurement temperature allows minimizing
thermal diffusion of defects. For low fluences, implantation defects
in GaN were shown to become mobile at temperatures around
120 K, while more stable defect complexes are formed for high
fluences suppressing any annealing for temperatures up to room
temperature [12].

RBS/C characterization was performed using a 1.4 MeV He⁺ ion
beam and backscattered particles were detected at 170°. Random
spectra were taken before and after each series of implantation by
tilting the sample by 5° and rotating the azimuthal angle. After each
implantation step, a channelling spectrum was taken for which the
samples were aligned so that the analysing beam coincided with
the growth axes for each of the GaN films i.e. <11̅20>, <0001>, and
<10̅10> for a, c, and m-plane material, respectively. The as-grown
samples showed RBS/C minimum yields close to the surface of
0.58(3), 0.75(3)% and 1.57(4)% for a-GaN, c-GaN and m-GaN,
respectively, revealing excellent single crystalline quality of all
layers along the growth direction.1Fig. 1 shows two-dimensional
(2D) maps of the normalized angular backscattering yield around
the three crystal directions simulated using the Monte Carlo code
FLUX [32]. The strong decrease of the backscattering yield for axial
and planar channelling directions is clearly visible. The theoretical
minimum yields along the main axes for a perfect crystal at 15 K
and using a Debye temperature of 446 K were estimated to be
0.36(5), 0.78(5)% and 1.01(8)% for a-GaN, c-GaN and m-GaN,
respectively. These values compare very favourably with the
experimental data showing only slightly increased values for the
cases of a-plane and m-plane material due to native defects in the
as-grown material.

Next, the difference in the minimum yield, Δxmin, between
the implanted and as-grown sample was determined for each channel
in the spectrum, using
Δxmin = (Yaült<Yran>)/Yran,
where Yaült and Yran are the yields in the aligned (al) spectra of the
implanted (impl) and as-grown (virgin) samples and Yran is the random yield.
In order to determine the relative concentration of displaced lattice
atoms versus depth, in the following called the damage profile or
relative damage level, the background due to dechannealling of the
analysing beam at implantation defects needs to be taken into ac-
count. This was done using a two-beam model which assumes two
fractions of the analysing beam particles, a random fraction that
interacts with all target atoms in the same way as for an amorphous
target and an aligned fraction which only interacts with displaced
lattice atoms (defects or thermally displaced atoms) [33–35]. In the
case of randomly displaced lattice atoms, the probability for an ion
to get dechannealled, i.e. to transit from the aligned to the random
beam, can be calculated and thus, the relative concentration of
placed lattice atoms versus depth (usually called defect profile)
can be extracted [36]. However, more complex defect structures,
namely extended defects such as dislocation loops and stacking
faults will influence the dechannealling of the analysing ions in a
different way. In this case the dechannealling probability cannot be
calculated but has to be adapted for proper subtraction of the
dechannelling background [37]. In our case this is done by
decreasing the critical angle of channelling. By doing so, we sepa-
rate direct backscattering due to displaced atoms from dechan-
nealling due to distorted channels caused by extended defects. As
will be shown below, for low fluences, this procedure results in a
very good agreement of our experimental results with SRIM Monte
Carlo simulations. Furthermore, the results agree well with similar
implantations and RBS/C measurements performed in c-GaN and
analysed using the Monte Carlo code McCasy [13,24]. Neverthe-
less, it should be kept in mind that the defect fractions extracted in
this way do not discriminate between the different defect

1 The minimum yield is the ratio of the yield of the aligned to that of the random
spectrum within a region of interest close to the surface and it serves as a quan-
titative measure of the crystal quality of a single crystal.
microstructures. In fact, extended defects such as dislocations and stacking faults contribute much less to the direct backscattering peak than randomly displaced atoms due to point defects and their clusters as well as amorphous zones [38].

Conventional Transmission Electron Microscopy (TEM) and high resolution TEM (HRTEM) were carried out ex situ on samples implanted to a fluence of $8 \times 10^{15}$ at/cm$^2$ and $4 \times 10^{16}$ at/cm$^2$. At these high fluences, RBS/C in c-GaN showed that annealing of implantation defects when heating the samples to room temperature is negligible [12]. TEM cross section specimens were prepared by mechanical polishing down to lower than a few μm using a multiprep™ machine, they were then glued on copper slot grids. The electron transparency was achieved by Ar ion milling at 5 keV using a GATAN precision ion polisher system (PIPS) at an incidence angle of $5^\circ$ while cooling the sample holder with liquid nitrogen. In order to further minimize the ion beam damage, a final milling step was carried out at $75^\circ$ incidence angle with the beam energy decreased to 0.5 keV. The TEM investigations were performed with a JEOL 2010 microscope and the HRTEM was carried out in a JEOL 2010 FEG instrument, both operated at 200 keV.

3. Results and discussion

3.1. Rutherford backscattering/channelling

Fig. 2 shows typical random and the aligned RBS/C spectra for a-GaN and m-GaN for the entire fluence range. Spectra for c-GaN, implanted and measured using the same conditions, are very similar to those shown for m-GaN and have been published previously [12,16]. As expected, an enhancement of the backscattering yield with increasing implantation fluence is observed as the number of displaced atoms increases with fluence. Several similarities are seen in the spectra for a-, c-, and m-GaN: All three materials show a bimodal damage distribution with a pronounced, sharp defect peak at the surface and another broader one deeper in the bulk crystal as previously reported for c-GaN [39]. Several regimes in the damage accumulation dynamics can be observed with fluence regions in which the backscattering yield, and thus the damage level, saturates and others in which a strong increase of backscattering yield is observed. For the highest fluences, the backscattering yield of the aligned spectra reaches the random yield. This happens at the surface as well as within a thin, buried layer deeper in the sample. In many semiconductors this is an indication for amorphisation, however, for c-GaN and implantation at room temperature it was shown that the highly damaged surface layer is rather due to the formation of a layer consisting of randomly oriented nanocrystallites [18]. We will show below that this is also....
the case for a- and m-GaN in the present implantation conditions. With still higher fluence the buried nanocrystalline layer grows in width, both toward the surface and to the bulk.

Finally, all spectra show high dechannelling yields in the aligned RBS/C spectra, that is, the backscattering yield remains high even for deep, unimplanted layers (at channel numbers below ~280). These high yields are due to the dechannelling of the analysing beam at defects and suggest the presence of extended defects similar to the results reported for c-GaN [12,13].

The main difference between the damaging dynamics of the three materials is seen at intermediate fluences (between $1 \times 10^{15}$ and $2 \times 10^{16}$ at/cm$^2$) where a-GaN shows considerably lower backscattering yields than c- and m-GaN.

In order to subtract the above mentioned dechanneling backgrounds and to allow a more quantitative discussion of the defect accumulation mechanisms the defect profiles were derived from the RBS/C spectra using a two-beam model of dechanneling as described above. Selected profiles, belonging to the five different regimes of damage build-up as indicated on the right side of each row, are shown in Fig. 3. The grey shaded areas correspond to the depth window used to plot the damage build-up curves in Fig. 4.

![Fig. 3. Defect profiles for a-GaN (left), c-GaN (middle) and m-GaN (right) derived from the RBS/C spectra using a two-beam model. Typical curves for selected fluences are shown which correspond to the five different regimes of damage build-up as indicated on the right side of each row. The grey shaded areas correspond to the depth window used to plot the damage build-up curves in Fig. 4.](image-url)
atomic density of GaN (8.8467 x 10^{22} atoms/cm^3). The distributions of vacancies and Ar as derived using the Monte Carlo code SRIM (version 2013) are also included in Fig. 3 for comparison.

Fig. 4 represents the average relative defect level in a region of interest from ~140 nm to ~170 nm depth (indicated by the grey shadowed area in Fig. 3). This depth window corresponds to the region where the aligned spectra first reach the random level. This choice permits the evaluation of the last step of nanocrystallisation. The damage build-up curves show a qualitatively similar behaviour for the three materials: Damage accumulation proceeds in five regimes (three steps separated by fluence regions in which damage saturates or increases at a very slow rate). For low fluences (between 2 x 10^{12} and 6 x 10^{13} at/cm^2) the relative damage level increases approximately linearly with implantation fluence (regime I). In regime II (between 6 x 10^{13} and 1 x 10^{15} at/cm^2) a first saturation of the damage level occurs followed by a steep increase in regime III (between 1 x 10^{15} and 4 x 10^{15} at/cm^2). Above 4 x 10^{15} at/cm^2 a second saturation occurs (regime IV) until finally a complete loss of the channeling effect is observed and the damage level saturates at ~100% (regime V). This last step occurs at slightly different fluences for the three materials revealing that nanocrystallisation occurs first in c-GaN (at ~1.2 x 10^{16} at/cm^2), then in m-GaN (at ~2 x 10^{16} at/cm^2), and finally in a-GaN (at ~3 x 10^{16} at/cm^2).

The described damage build-up curves are typical for materials with strong dynamic annealing effects which lead to efficient recombination of point defects already during the implantation [40]. In fact, similar curves have been reported by many groups for implantation in c-plane GaN, AlN as well as AlGaN thin films and nanostructures at room temperature and cryogenic temperatures [11–13,22,41–44]. For c-GaN, the derived fractions of displaced atoms agree very well with a similar study where Monte Carlo simulation was used to discriminate between direct backscattering due to randomly displaced atoms and dechanneling due to crystal distortions caused by extended defects [24,13].

For many semiconductors, including c-GaN and AlGaN [22], this defect dynamics was successfully described by the three-step amorphisation model by Hecking which takes into account defect formation and recombination mechanisms [45,40]. In the present case, nanocrystallisation is considered instead of amorphisation maintaining the same mathematical description. In fact, amorphisation and nanocrystallisation cannot be distinguished in the RBS/C spectra since both lead to a complete breakdown of the channeling effect. The model is based on two coupled differential equations for both point defect formation and nanocrystallisation/amorphisation, described by [45,40]:

\[
\frac{dn_{pd}}{d\Phi} = P_{pd}e^{-\frac{R_{pd}}{C_{pd}}}(1 - n_{nc}) + C_{pd}n^{1.2}_{pd}\left[1 - \frac{n_{pd}}{n^* (1 - n_{nc})}\right]
\]

\[
\frac{dn_{nc}}{d\Phi} = \frac{n_{pd}}{1 - n_{nc}}
\]

\[
\frac{dn_{nc}}{d\Phi} = (P_{nc} + G_{nc}n_{nc})(1 - n_{nc}).
\]

In Eq. (1), \(n_{pd}\) and \(n_{nc}\) are the relative concentration of point defects (and their clusters) and the nanocrystalline fraction, respectively, so that the total fraction of displaced atoms is given by \(n_{tot} = n_{pd} + n_{nc}\). At low fluence, the defect accumulation dynamics is controlled by \(P_{pd}\) (the cross section for point defect formation) and \(R_{pd}\) (the point defect recombination rate). \(C_{pd}\) describes the formation of non-recombining point defect clusters with a saturation concentration \(n^*\). The formation of nanocrystalline regions depends on \(P_{nc}\) (the cross section for the formation of nanocrystalline material) or seeds for nanocrystallisation) and \(G_{nc}\) (a measure of the strength of the stimulated growth of nanocrystalline regions) [40]. Eq. (1) was solved numerically and the six parameters of the model were obtained by iterative fitting of the experimental data in the different regimes starting at low fluences. Fits to the experimental data using this model are shown in Fig. 4 as straight lines and the derived fitting parameters are summarized in Table 1.

It is clear from the damage build-up curves in Fig. 4, as well as the similar fitting parameters \(P_{pd}\) and \(R_{pd}\), that the first step of damage production is very similar for all three orientations. The defect formation cross section \(P_{pd}\) describes the linear increase of the damage build-up curve in regime I which is attributed to the formation of point defects within well separated collision cascades caused by ions hitting pristine material. The fact that the defect formation cross section is the same for the three materials suggests that firstly, there is no pronounced anisotropy of displacement energies for the three orientations and secondly, the created point defects are randomly distributed within the lattice. For displacements along defined directions, shadowing effects would occur, i.e. defects would be shadowed by the atomic rows during RBS/C measurements along certain directions which would affect the apparent defect level measured by RBS/C. It is also worth to mention that the value \(P_{pd}\) derived here is in good agreement with previous estimations of the defect formation cross section in c-GaN and in perfect agreement with the value estimated from SRIM simulations [25].

In regime II, collision cascades start to overlap leading to an increased recombination of point defects resulting in a first saturation of damage level. The recombination rate, described by \(R_{pd}\), is similar for c-GaN and a-GaN but slightly lower for m-GaN leading to a slightly higher saturation value for \(n_{sat}\) in the latter case (the saturation level is approximately given by the quotient \(P_{pd}/R_{pd}\)). This lower capacity for point defect recombination is most likely attributed to the as-grown material may reduce the diffusivity of point defects.

For higher fluences and implantation defect levels in regime III and beyond, this effect should be negligible. In fact, for c-GaN implanted to low fluences (regime I) it was shown that defects retained after implantation at 15 K become mobile around 120 K while for higher
fluctuations (at the beginning of regime III) defects are stable up to room temperature [12]. The defect profiles for regimes I and II agree well with the vacancy profiles predicted by SRIM simulations (see Fig. 3) which indicates that for these low fluences, ballistic effects due to nuclear interaction between the ion and the host atoms and the subsequent collision cascade are dominant for the defect formation.

In regime III, the defect profiles start to deviate from the shape predicted by SRIM simulations. A damage peak in the bulk grows fast with increasing fluence. For c-GaN it is placed close to the maximum of nuclear energy deposition while for a-GaN and m-GaN it is slightly deeper suggesting that beside the primary defects an influence of the Ar-ions themselves cannot be discarded. The strong increase of maximum damage level occurs in the same fluence region for the three orientations but it is steepest for c-GaN (Fig. 4) which is also seen in the higher value of the fitting parameter \( C_{pd} \) describing the formation of defect clusters (Table 1). At the same time, in the region between the surface and bulk defect peaks, the defect concentration remains very low in contrast to the SRIM simulation (Fig. 3, regime III). This deviation from the expected defect profile suggests efficient defect diffusion towards the surface and the bulk peak where point defects probably get trapped at extended defects leading to their growth. In fact, detailed TEM investigation of rare earth implanted c-GaN revealed, for a fluence range corresponding to regime III, the formation of a dense network of basal and prismatic stacking faults [14,18]. This network starts to form in the bulk and then gradually extends towards the surface. It was suggested that this network acts as a diffusion path for point defects which then accumulate at the surface and lead to the formation of the nanocrystalline surface layer found for room temperature implantation in c-GaN [14]. Therefore, the deviation of the defect profile from the expected form, the strong increase of maximum defect density with fluence as well as the high dechanneelling yields found from the RBS/C spectra for regime III point to a change of defect nature from dominantly point defects and small clusters to larger defect clusters and extended defects.

In regime IV, the maximum concentration of displaced atoms saturates while the defect profiles seen are broaden englobing now the entire implanted region. In the case of a-GaN, the defect profiles even seem to extend beyond the implanted region suggesting the diffusion of defects into pristine material. Although care should be taken because the data treatment for dechanneelling correction may lead to artificial tails at the deeper end of the defect profile if the model used is not adequate for the actual defect morphology, the wider defect distribution in a-GaN was confirmed by TEM images as will be shown below.

The most remarkable difference observed for the three materials is the significantly lower value of the defect saturation level \( n^* \) which is only 12% for a-GaN and considerably higher for c-GaN (58%) and m-GaN (52%). A similar effect has been reported before for a-GaN implanted with Eu ions [26]. However, from RBS/C measurements alone it is difficult to decide if it is really the defect density or defect type which changes or if the same defect configuration leads to different backscattering yields along certain crystallographic directions due to shadowing or steering effects. In fact, the distributions of the analysing ions in the channel for a certain energy at a certain depth may influence the sensitivity of RBS/C to certain defect microstructures [46].

Anisotropies of implantation damage for irradiation along different crystallographic directions have been discussed for several crystals such as ZnO [47], YSZ [48,49], MgO [48,50], \( \text{Al}_2\text{O}_3\) [51,52] and \( \text{LiNbO}_3\) [53]. However, in some materials it was demonstrated that such anisotropies can be due to the measurement procedure and not caused by actual differences in damage formation [49,53]. Among the cases mentioned above, that of ZnO is particularly interesting since ZnO crystallizes in the wurtzite lattice structure like GaN and both materials have strong similarities regarding bandgap, radiation hardness and dynamic annealing effects [12]. In particular, similar defect structures, namely extended defects such as stacking faults and dislocation loops are formed upon implantation [54]. Similar to our observations in GaN, Charnvanichborikarn et al. report a significantly lower RBS/C yield at the bulk damage peak of a-ZnO as compared to c-ZnO and an only slightly lower damage level for m-ZnO [47].

Besides the different visibility of the same defect constellation from different directions there are several “real” effects that can cause such asymmetry: 1) Different displacement threshold energies for different directions may cause a higher damaging potential for irradiations along certain crystal directions. Indeed, a small anisotropy in displacement energies was predicted for GaN by molecular dynamics simulations [55], however, the fact that damage profiles are similar for low fluences (regimes I and II) proves that these effects are not significant for the dense collision cascades caused by ion implantation. This is in agreement with measurements of implantation damage in other materials [49,53].

2) Diffusion of point defects may be different in the presence of different surfaces. Indeed, for all three orientations, pronounced surface damage peaks suggest that the surface acts as a sink for migrating point defects. It is clear that the surface peak is more pronounced in a-GaN while the defect level between bulk and surface peak is much lower in this material suggesting that defect diffusion to the surface is more efficient than in c- and m-GaN. At the same time the bulk damage level remains very low suggesting that different kinds of defect complexes are formed which are less efficient in trapping new point defects or they are trapped in positions where they do not contribute to the direct backscattering (non-random positions). In fact, the defect profiles in a-GaN seem to reach deeper into the crystal than for the other two materials (see Fig. 3 regime IV). 3) The mechanisms discussed above may be further influenced by the different surfaces i.e. due to different sputtering yields or stoichiometric changes due to selective sputtering of nitrogen.

In order to understand the pronounced differences in damage level in regime IV, additional samples were implanted to a fluence of \( 8 \times 10^{15} \text{ at/cm}^2 \) for ex situ TEM analysis as will be discussed below.

The last step of damage accumulation (regime V) is again similar for the three materials although it occurs at slightly different fluences: first in c-GaN, then in m-GaN and last in a-GaN as mentioned above. This step is represented by the parameters \( P_{nc} \) and \( G_{nc} \) (see

<table>
<thead>
<tr>
<th>Material</th>
<th>( P_{pd} ) (( 10^{15} \text{ cm}^2 ))</th>
<th>( R_{pd} ) (( 10^{15} \text{ cm}^2 ))</th>
<th>( C_{pd} ) (( 10^{15} \text{ cm}^2 ))</th>
<th>( n^* )</th>
<th>( P_{nc} ) (( 10^{15} \text{ cm}^2 ))</th>
<th>( G_{nc} ) (( 10^{15} \text{ cm}^2 ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>a-GaN</td>
<td>0.7</td>
<td>22</td>
<td>1.6</td>
<td>0.12</td>
<td>2</td>
<td>6</td>
</tr>
<tr>
<td>c-GaN</td>
<td>0.7</td>
<td>22</td>
<td>2.2</td>
<td>0.58</td>
<td>2</td>
<td>13</td>
</tr>
<tr>
<td>m-GaN</td>
<td>0.7</td>
<td>15</td>
<td>1.5</td>
<td>0.52</td>
<td>2</td>
<td>8</td>
</tr>
</tbody>
</table>
Table 1). Test calculations with Eq. (1) show that these parameters are not very well defined. Correlated variations of the two parameters may yield similarly good representation of the experimental data (e.g. $P_{nc} = 2 \times 10^{-29}$ cm² and $G_{nc} = 13 \times 10^{-16}$ cm² yield equally good fits for the curve of a-GaN in Fig. 4). Despite this, it is clear that in any case $P_{nc}$ is too small to represent a cross section for

Fig. 5. Cross sectional TEM images of c-GaN (a,b), a-GaN (c,d) and m-GaN (e,f) implanted to a fluence of $8 \times 10^{15}$ at/cm². The defect profiles derived by RBS/C were superimposed to the TEM images. In d) a large density of dislocation loops can be observed, some of which were marked by yellow arrows. In f) some planar defects are marked with blue arrows and dislocation loops with yellow arrows. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
direct impact formation of amorphous or nanocrystalline material. Instead $P_{nc}$ acts to emulate the nucleation of this type of damage which then grows during further implantation. As seen in Fig. 3, a complete loss of channelling effect (either due to amorphisation or due to nanocrystallisation) is observed in a very thin buried layer at a depth that neither corresponds to the maximum of nuclear energy deposition nor to the maximum of the Ar-profile. It seems that both defects and impurities are necessary to nucleate this layer. The width of this highly damaged layer then increases with increasing fluence and this stimulated growth seems to be faster towards the surface than towards the bulk, probably due to the lower density of primary defects in the deeper layers. However, even for the highest fluence, a thin layer of single crystalline material remains between the surface and the bulk damage peaks.

3.2. Transmission electron microscopy

TEM cross sectional images of the entire implanted layer for samples implanted to a fluence of $8 \times 10^{15}$ at/cm² are shown in Fig. 5 for c-GaN (a,b), a-GaN (c,d) and m-GaN (e,f). The RBS/C defect profiles superimposed to these images agree well with the defect contrast observed in the TEM images for all materials. Fig. 5 a, c and e show images in 0002 weak beam conditions which reveal displacements along the [0001] direction, that is the c-axis of the wurtzite lattice. Such displacements can for example be caused by point defect clusters. A clear bright contrast can be seen for c and m-GaN and almost nothing particular along [0001] is visible in a-GaN as attested by the presence of thickness fringes which have not been disturbed by the implantation damage. This may suggest a different defect generation process during the implantation in GaN along the a direction.

In the case of c-GaN, when images are taken in $g = 10\bar{2}0$ conditions (Fig. 5b), a typical contrast of bright lines parallel to the surface is observed. Similar results have been obtained by several groups for ion implanted c-GaN and arise from the formation of a dense stacking fault network consisting mainly of $I_1$ basal stacking faults which are interconnected by prismatic stacking faults [10,14,17]. High resolution images (not shown) confirm that such a network is indeed formed and reaches from the surface to a depth of ~280 nm. Images taken in the same condition in a-GaN (not shown) and m-GaN (Fig. 5f) reveal a number of growth-related stacking faults on the c-plane which are the dominant defects created during heteroepitaxial growth of non-polar material; typically they originate at the interface with the substrate and propagate up to the surface [56]. In the implanted region these stacking faults are interrupted by the implantation damage and no additional basal stacking faults are formed in the implanted region of a-GaN in contrast to c-GaN. The different nature of extended defects created by the implantation is clearly seen in Fig. 5f showing an image of a-GaN in $g = 112\bar{0}$ conditions. In a layer starting about 10 nm below the surface and extending to about 250 nm depth, a large density of linear defects, most probably dislocation loops with a c $\times 112\bar{0}$ character, has formed. Beyond this area, a defect contrast due to displacements along the $<112\bar{0}>$ direction is also visible with an extension until ~400 nm confirming that the defect profile after implantation in a-GaN runs deeper into this sample than for c- and m-GaN as it was already indicated by the RBS/C measurements. For the case of m-GaN, Fig. 5e recorded with $g = 0002$ weak beam condition shows two typical defect systems: i) the partial dislocations associated to the stacking faults coming from the as-grown bulk GaN part which continue to the surface through the implanted region. ii) a more irregular white contrast inside the implanted area from the deformation along the c-axis that is due to defect clusters. In Fig. 5f, whereas the density of bulk stacking faults is comparable to that of the partial dislocations of Fig. 5e, many more features which are mainly vertical can be seen in the implanted zone. This is a clear indication that additional planar defects have been generated during the ion implantation but also dislocation loops are visible. In fact, the defect microstructure has similarities with that of both c-GaN and a-GaN. Therefore as seen in these two figures, the implanted area exhibits a highly increased density of defects which agrees with the high fraction of displaced atoms evidenced by RBS/C. Fig. 6 shows TEM images for a- and m-GaN after the implantation to the highest fluence of $4 \times 10^{16}$ at/cm². In both cases, a nanocrystalline layer is formed at the surface and another one in the bulk both separated by a thin single crystalline layer marked by stars in Fig. 6. The high resolution images show details of the interface area between this single crystalline interlayer and the adjacent nanocrystalline material. Well aligned lattice fringes confirm the single crystalline character of the interlayer in contrast to the nanocrystalline material where a number of small areas with randomly oriented lattice fringes are resolved. The buried nanocrystalline layer furthermore shows a large number of circular bubbles or voids. Their diameter is largest close to the end of range of the implanted argon ions around 200 nm suggesting that Ar bubbles are formed at this depth. However, similar features were observed in Au-implanted c-GaN attributed to the dissociation of GaN and the formation of N-bubbles [57]. In the present case, most probably gas filled bubbles contain both Ar and N.

A good qualitative agreement between RBS/C and TEM is observed; the nanocrystalline layers lead to a complete overlap of random and aligned spectra while the single crystalline interlayer is clearly visible as a dip in the aligned spectra. However, a direct superposition of the RBS/C defect profiles with the TEM images (not shown) reveal a discrepancy in the depth scale. This discrepancy is due to the change of GaN density upon high fluence irradiation. Note that the density of pristine GaN (6.15 g/cm³) was used to convert the RBS depth scale, measured in at/cm², to length units what leads to an underestimation of any layer thickness if the density is reduced by the irradiation. In fact, the volume expansion of c-GaN upon ion implantation is well documented in the literature [10,57]. A good agreement between TEM and RBS/C defect distribution is achieved when a decrease of the GaN density by 23% is assumed which is in line with published data on c-GaN [57]. Very similar TEM results have also been reported for room temperature Ar implantation into c-GaN [24]. We can therefore conclude that the defect microstructure and distribution in regime V is very similar for polar and nonpolar films. A slight difference is seen in the width of the single crystalline interlayer which is wider in a-GaN than in m-GaN. This is most probably an effect of the stronger dynamic annealing in a-GaN which leads to lower defect levels in a depth region between the surface and the bulk damage peak and which also explains the delay in the on-set of nanocrystallisation (regime V of the damage build-up curve is reached at higher fluences in a-GaN, see Fig. 4). Another difference which is worth to mention is the very smooth surface of m-GaN for this high fluence while a-GaN presents increased roughening of the surface.

4. Conclusions

A detailed study of damage formation upon 300 keV Ar implantation at 15 K has been performed in a-, c- and m-plane GaN combining in situ RBS/C and ex situ TEM. Damage accumulation proceeds in three steps which can be well described using the defect formation and recombination model proposed by Hecking et al. [45]. The first step is characterized by an approximately linear increase of the defect concentration with fluence followed by a
fluence region were defect recombination dominates and the defect level saturates. The defect formation and recombination rates for these low fluences are very similar for the three crystal orientations showing that anisotropies in displacement threshold energies along the main crystallographic directions are negligible for the present implantation conditions and that displaced atoms are mostly distributed randomly.

The second step is attributed to the formation of defect clusters and extended defects leading to a deviation of the defect profiles from the expected shape and a strong increase of the concentration of displaced atoms followed by a subsequent saturation. In this fluence range, pronounced differences are seen for the three crystal orientations. In particular, the defect saturation level is more than 4 times lower in a-GaN (12%) than in c-GaN (58%) and m-GaN (52%). Furthermore, the microstructure of extended defects is very different in this fluence range consisting of a network of basal stacking faults in the case of c-GaN and a high concentration of dislocation loops with <11\overline{2}0> character for a-GaN. In m-GaN, a large density of extended defects is created in the implanted region and the defect microstructure resembles that of planar defects like in c-GaN mixed with dislocation loops as in a-GaN. However, the defect levels measured by RBS/C are nearly as high as for c-GaN pointing to the formation of a large density of point defect clusters besides stacking faults and dislocation loops.

The complete loss of the channeling effect is observed in all crystals at fluences around 1–3 \times 10^{16} \text{ at/cm}^2 occurring first in c-GaN, then in m-GaN and finally in a-GaN. This loss of single crystalline order is due to the formation of two nanocrystalline layers, one at the surface and one buried at a deeper depth and containing gas bubbles or voids. The position of the buried nanocrystalline layer is found to start between the depths of maximum nuclear energy deposition and maximum Ar-concentration and then this layer grows in width for increasing fluence.

In conclusion, we showed that a-plane GaN is more resistant to radiation damage than c- or m-plane material in a fluence range from 1 \times 10^{15} to 3 \times 10^{16} \text{ at/cm}^2. This is attributed to different dynamic annealing processes in the vicinity of these surfaces which lead to very distinct defect microstructures. More work is needed to understand the driving forces that lead to the transformation between distinct defect microstructures in the various fluence range.

Fig. 6. Cross sectional TEM images of a-GaN and m-GaN implanted to a fluence of 4 \times 10^{16} \text{ at/cm}^2. For both orientations high fluence implantation causes the formation of a buried nanocrystalline layer containing voids/bubbles and a nanocrystalline surface layer. Between these highly damaged layers a thin single crystalline interlayer is clearly visible (marked by a star). The high resolution images show details at the interface between this single crystalline interlayer (darker contrast) and the nanocrystalline material (light contrast) clearly showing randomly oriented lattice fringes in the latter.
regimes. Catarino et al. observed a strong expansion of the a-plane parameter in the implanted volume of a-GaN [26]. Considering that the present work was performed at 15 K where thermal effects should be small, it is likely that stresses introduced by the implantation defects contribute to the defect transformations, as it has been suggested for c-GaN [24,21]. In contrast to c-GaN where such stresses lead to a homogeneous compressive strain in the growth plane, for a-GaN this will lead to anisotropic strain [58] within the implanted volume which may contribute to the formation of distinct defect microstructures at intermediate fluences. Also the effect of these different extended defects on electrical or optical properties needs to be understood when ion implantation is used for device processing.

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References


