High-quality bulk *a*-plane GaN sliced from boules in comparison to heteroepitaxially grown thick films on *r*-plane sapphire

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Thick GaN bars with [11$\bar{2}$0] orientation have been sliced from GaN boules grown on freestanding films by hydride vapor phase epitaxy (HVPE) in the [0001] direction. High-resolution x-ray diffraction and transmission electron microscopy have been used to study the structural quality and defect distribution in the material in comparison to heteroepitaxially grown thick HVPE-GaN films grown in the [11$\bar{2}$0] direction on (1$\bar{1}$02)-plane sapphire. It is demonstrated that while the heteroepitaxial material possesses a high density of stacking faults and partial dislocations, leading to anisotropic structural characteristics, the (11$\bar{2}$0)-plane bulk GaN, sliced from boules, exhibits low dislocation density and narrow rocking curves with isotropic in-plane character. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2236901]

Over the last few years, considerable attention has been paid to the development of nitride epitaxial layers and heterostructures with nonpolar and semipolar surface orientations, driven by the need to avoid the built-in electric fields, hampering the performance of the nitride-based devices grown in the conventional [0001] direction. Different growth techniques, substrate materials, and substrate and layer orientations have been suggested and investigated.[1–9] Practically all the publications consistently show that in addition to the well known problems for heteroepitaxially grown nitrides in the [0001] direction on sapphire and SiC, such as high dislocation density and strain induced effects, the material grown in the [11$\bar{2}$0] direction on both (1$\bar{1}$02)-plane sapphire and (11$\bar{2}$0)-plane SiC also experiences a high density of stacking faults and in-plane anisotropy of all the properties, imposing more difficulties on the measurements and analyses. Lower mismatch substrates such as $LiAlO_3$ and $MgAl_2O_4$ have been suggested with the hope to enable lower defect density, but still similar structural and impurity related problems have been observed.[8,9] The well known approach for lateral epitaxial overgrowth, employing stripe patterns, has also been used in *a*-plane growth and has been found to improve the morphology and to reduce significantly the defect density, but still the optical properties have been defect dominated.[2,3,5] Hydride vapor phase epitaxy (HVPE) growth of GaN layers, proven to be capable of producing very thick layers with significantly improved quality along the [0001] growth direction, has not been so successful for layers grown in the [11$\bar{2}$0] direction,[5–7] clearly due to the specific type and distribution of defects in this direction.

An alternative would be a material grown in the conventional [0001] direction up to a significant thickness and then a slicing of bars and/or wafers with a desired nonpolar orientation. At present, the only promising technique for producing nitride boules thick enough to allow slicing of reasonable large areas is the HVPE growth. In this work, we carry out a comparative study of the structural properties, as determined by high-resolution x-ray diffraction (HRXRD) and transmission electron microscopy (TEM), of (11$\bar{2}$0)-plane GaN grown by HVPE either in [11$\bar{2}$0] or in [0001] directions.

Heteroepitaxial growth of GaN films in the [11$\bar{2}$0] direction on (1$\bar{1}$02)-plane sapphire was carried out at Linköping University in a conventional horizontal reactor at 1080 °C on metal organic vapor phase epitaxial GaN template layers.[6] The templates of about 1.2 μm in thickness, including SiN micromasks for threading dislocation reduction, were grown at Bremen University in a Thomas Swan vertical reactor.[10] Samples with different thicknesses (5–90 μm) have been studied but the structural properties and defect distributions have been found similar. The second type of GaN material was grown by HVPE at Kyma Technologies. A homoepitaxial growth was conducted on a freestanding GaN film also produced by HVPE and separated from the sapphire. Boules with 2 in. diameter and thickness up to 7–8 mm were grown in the [0001] direction. Then rectangular bars with sizes of 10×6 mm^2 have been sliced parallel to the (11$\bar{2}$0) plane.

The structural properties of the samples were investigated by HRXRD at room temperature using Philips x-ray diffractometer equipped with a parabolic graded x-ray mirror collimator followed by a twofold monochromator and a three-crystal analyzer. The spot size has been 1×12 mm^2 unless other conditions are specified for a specific experiment. Reciprocal space maps (RSMs) of the 11$\bar{2}$0 reflection were recorded as consecutive 2θ-ω scans each separated by an ω offset. TEM was carried out on a high-resolution transmission electron microscope operated at 200 kV. Plan-view TEM specimens were prepared by mechanical polishing followed by Ga ion milling in a Nova Nanolab (FEI) dual beam focused ion beam (FIB) system operated at 30 kV.

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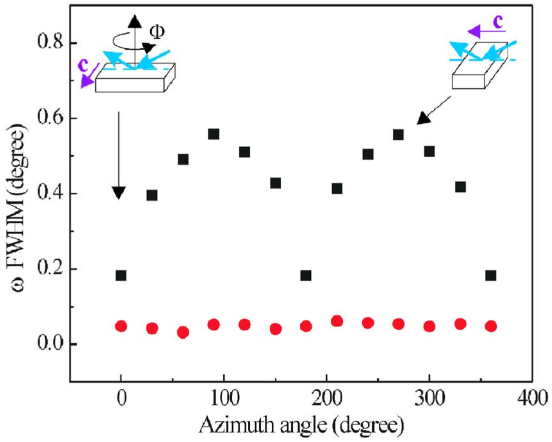

FIG. 1. Azimuth angle dependence of the FWHM of the $11\bar{2}0$ rocking curves for a heteroepitaxially grown *a*-plane GaN film in the [$11\bar{2}0$] direction (square symbols) and an *a*-plane GaN bar sliced from GaN boule grown in the [0001] direction (dot symbols).

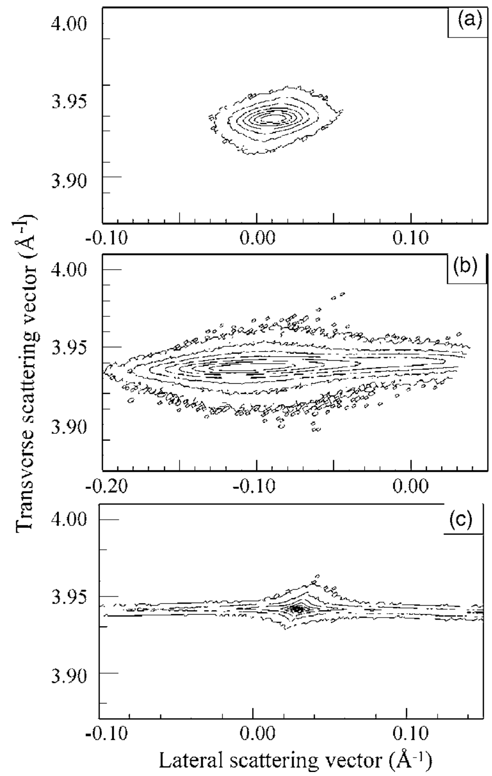

FIG. 2. Reciprocal space maps around the $11\bar{2}0$ reciprocal point of [(a) and (b)] an *a*-plane GaN film grown by HVPE on *r*-plane sapphire with a MOCVD-GaN template layer for scattering plane (a) parallel and (b) perpendicular to the GaN basal plane and (c) an *a*-plane GaN bar sliced from HVPE grown GaN boule, being independent of the sample orientation with respect to scattering plane.

Aiming to reveal a possible in-plane anisotropy of the structural characteristics we analyzed the rocking curves (RCs) of the main $11\bar{2}0$ reflection at symmetric diffraction geometry as a function of the azimuth angle for sample rotation over a full circle range. Figure 1 shows the azimuth dependences of the full width at half maximum (FWHM) of the RCs for the two samples. An M-shape dependence (square symbols) of the heteroepitaxially grown *a*-plane GaN sample is revealed with a difference by a factor of 3 between the minimum value of 0.18° (when the scattering plane is parallel to the GaN basal plane) and the maximum value (when the scattering plane is perpendicular to the GaN basal plane). Such a type of in-plane anisotropy is in agreement with the results reported for metal organic chemical vapor deposition (MOCVD) grown *a*-plane GaN with/without stripe-patterned template,[3] for HVPE grown *a*-GaN employing AlN buffer layer,[7] as well as for HVPE grown GaN films without a buffer.[11] Interestingly, the range of the FWHM between 0.2° and 0.6° over a 360° range is almost independent of the growth technique and the nucleation scheme/optimization used. The latter suggests that such an anisotropic behavior is specifically characteristic for the anisotropic nature of the growth in this direction, most likely related to unavoidable defects with a specific distribution. In contrary, the azimuth dependence of the RC width (dot symbols) of the *a*-plane GaN bar sliced from a boule shows an isotropic behavior with a value of about 90–160 arc sec, being almost an order of magnitude lower than the best values for the heteroepitaxial material. The latter is indicative of a higher structural quality of the material grown in the conventional [0001] direction and sliced along a nonpolar surface.

In order to reveal the reasons for the RC broadening behavior, we recorded RSMs around the $11\bar{2}0$ reciprocal point. Figures 2(a) and 2(b) show the maps of the heteroepitaxially grown sample for two azimuth positions of the sample with respect to the scattering plane (as shown schematically in the insets of Fig. 1), namely, for the positions when the FWHM takes the minimum and the maximum values, respectively. While the first map is symmetrical and shows a relatively narrow main peak (FWHM of 0.18°), the second map is significantly broadened in the lateral direction, and in addition a low intensity asymmetry appears. A detailed study of the RC shape shows that the scan has a complex structure, and it contains two peaks that show substantially different behaviors upon azimuth variation. The first of them has an almost constant peak position, while the second peak position varies around the first peak within a range of about 4°, when the sample is rotated around its normal between 0° and 360°. In addition, the constant peak is most pronounced when the scattering plane is parallel to the GaN basal plane, while the second peak appears for azimuthal positions at which the scattering plane is not perpendicular to the GaN *c* axis. This clearly indicates that several factors contribute to the RC width with different weights, depending on the sample orientation relative to the scattering plane. In principle, in the case of the symmetric geometry, the RCs could be broadened by dislocation amount, mosaic tilt, small lateral coherence lengths of the mosaic blocks, and bending.[12] In order to check out for the different contributions we investigated the ω scans versus spot width size varied from 0.3 to 1 mm. We have found that the ω scan broadens by about 10% when the sample basal plane is perpendicular to the scattering plane, while the two peaks forming the ω scan when the sample basal plane is parallel to the scattering plane broaden by 25%–30% as determined from fitting procedure. This indicates a contribution by curvature and/or spread of mosaic blocks with sizes of the order of the beam size. The latter could be neglected as it leads to a broadening of the RCs below the resolution of the analyzer. The effect of the curvature on the RCs was additionally confirmed by shifting the beam spot on the sample surface either along the *c* direction or perpendicular to it. In both cases peak shifts were observed. We have estimated the curvature to differ by a factor of 3 in the two perpendicular in-plane

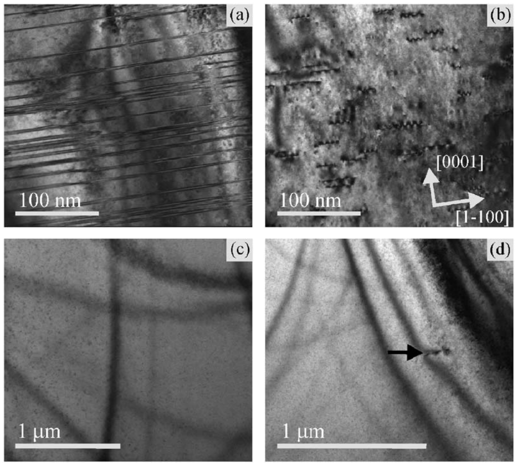

FIG. 3. [(a) and (b)] Bright-field plan-view TEM images of an *a*-plane GaN film grown on *r*-plane sapphire taken close to the $[11\bar{2}0]$ zone axis with (a) tilting by 4° towards the 0002 reflection for visualizing basal plane stacking faults and (b) tilting by 5° toward the $1\bar{1}00$ reflection for visualizing dislocations with a nonzero [0001] component. [(c) and (d)] Bright-field TEM images taken under similar conditions as for (a) and (b), respectively, for the *a*-plane GaN bar sliced from HVPE grown GaN boule grown in the [0001] direction. The arrow in image (d) marks the presence of an edge dislocation (as determined from weak-beam TEM analysis), which was the only one found in the investigated area of $30 \times 30\ \mu m^2$.

directions, having a maximum value of 2.5 m^{-1} in the [0001] direction, by measuring the RC positions at two points separated by a certain distance on the sample surface.[13]

A presence of mosaicity and defects has been investigated by plan-view TEM. Figure 3(a) shows a bright-field image of the *a*-plane GaN sample grown by HVPE on *r*-plane sapphire taken at a sample tilt of 4° towards the 0002 reflection, revealing a stacking fault density in the range of $10^5\ cm^{-1}$. In Fig. 3(b) a similar specimen area is shown in bright field with a tilt of about 5° towards the $1\bar{1}00$ reflection showing dislocation contrast, which was analyzed under weak-beam conditions and found to be stemming from Frank-Shockley-type partial dislocations (with Burgers vectors $\mathbf{b}=1/6\langle 02\bar{2}3\rangle$) bounding the basal plane stacking faults. These dislocations running in the $[11\bar{2}0]$ direction with a Burger vector component parallel to the GaN [0001] direction and density in the range of $10^9\ cm^{-2}$ are most likely to be formed at grain boundaries. These results indicate a contribution by both dislocation amount and small lateral coherence lengths to the large FWHM of the rocking curve, which is even more enhanced by the effect of curvature. The change of the subpeak position and intensity in the RC scans when the GaN is rotated at different angles with respect to the scattering plane could be related to a possible presence of a mosaic tilt and/or the specific distribution of the partial dislocations, whose strain fields are anisotropic due to the anisotropy of the wurtzite structure, and thus is reflected by different weights in the ω scans at different azimuths.

Figure 2(c) shows one representative RSM around the $11\bar{2}0$ reciprocal point of the *a*-GaN bar sliced from the boule, as being similar for all azimuth angles. In contrary to the complicated behavior of the RCs for the material grown in the $[11\bar{2}0]$ direction, the bulk material grown in the [0001] direction exhibits a single peak RC invariant to beam shift, beam spot size, and scattering plane orientation with a very narrow ω rocking curve of about 90 arc sec due to the finite lateral dimensions of the perfect crystal. The latter is confirmed by the plan-view TEM images [Figs. 3(c) and 3(d)] recorded under the same imaging conditions as for Figs. 3(a) and 3(b). Beside the contrast resulting from specimen bending no basal plane stacking faults are observed, and only one threading dislocation could be found in the investigated area of $30 \times 30\ \mu m^2$, resulting in a dislocation density in the range of $10^5\ cm^{-2}$ and below. A detailed analysis of the RSM in Fig. 2(c) also shows a lateral broadening at the tails of the ω curve. The latter could be explained by a contribution of diffuse scattering due to the dislocation strain field. Such a diffraction profile, consisting of two components, namely, a strong coherent scattering from regions of the perfect crystal between dislocations and diffuse scattering from regions around dislocations, is characteristic for high-quality material with low dislocation density.[14,15]

In summary, we have studied in a comparative way the structural quality of *a*-plane GaN grown heteroepitaxially in the $[11\bar{2}0]$ direction on *r*-plane sapphire and homoepitaxially grown gaN in [0001] directions subsequently sliced along the *a* plane. We have shown that the anisotropic nature of the $(11\bar{2}0)$ grown surface leads to anisotropic structural characteristics, manifested in asymmetry of the rocking curves due to contribution by the anisotropic partial dislocation distribution, mosaic tilt, small lateral coherence lengths of the mosaic blocks, and anisotropic bending in the two perpendicular in-plane directions. Instead, the *a*-plane bars sliced from GaN boules grown in the [0001] direction exhibit a low dislocation density with uniform in-plane distribution, manifested in narrow RSM around the $11\bar{2}0$ reciprocal point with diffuse scattering. The superior structural quality determines the conventional growth in the *c* direction with subsequent slicing in the direction of interest as the most promising approach for producing GaN substrates with nonpolar orientation.

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