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In-situ NC-AFM measurements of high quality AlN(0001) layers grown at low growth rate on 4H-SiC(0001) and Si(111) substrates using ammonia molecular beam epitaxy

Florian Chaumeton, Sébastien Gauthier, and David Martrou
CNRS / CEMES / GNS, 29 rue Jeanne Marvig, 30155 Toulouse, France

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Nitride wide-band-gap semiconductors are used to make high power electronic devices or efficient light sources. The performance of GaN-based devices is directly linked to the initial AlN buffer layer. During the last twenty years of research on nitride growth, only few information on the AlN surface quality have been obtained, mainly by ex-situ characterization techniques. Thanks to a Non Contact Atomic Force Microscope (NC-AFM) connected under ultra high vacuum (UHV) to a dedicated molecular beam epitaxy (MBE) chamber, the surface of AlN(0001) thin films grown on Si(111) and 4H-SiC(0001) substrates has been characterized. These experiments give access to a quantitative determination of the density of screw and edge dislocations at the surface. The layers were also characterized by ex-situ SEM to observe the largest defects such as relaxation dislocations and hillocks. The influence of the growth parameters (substrate temperature, growth speed, III/V ratio) and of the initial substrate preparation on the dislocation density was also investigated. On Si(111), the large in-plane lattice mismatch with AlN(0001) (19%) induces a high dislocation density ranging from 6 to 12×10^10/cm^2 depending on the growth conditions. On 4H-SiC(0001) (1% mismatch with AlN(0001)), the dislocation density decreases to less than 10^10/cm^2, but hillocks appear, depending on the initial SiC(0001) reconstruction. The use of a very low growth rate of 10 nm/h at the beginning of the growth process allows to decrease the dislocation density below 2×10^9/cm^2.

I. INTRODUCTION

During the last four decades, the global demand for electrical energy has been multiplied by 3.3 and is growing faster than for all other forms of energy. A solution to meet this demand is to integrate devices with greater operating high-powers, frequencies and temperatures. Wide band gap group-III nitride semiconductors are now widely used for the elaboration of electronic devices with such capacities. But the physical properties of nitride films are highly influenced by their defect density. Thus in order to obtain high performance devices, it is imperative to reduce this defect density as much as possible. The elaboration of nitride semiconductor devices is very often started with an aluminum nitride (AlN) buffer layer, which can be grown on silicon (Si(111)), silicon carbide (nH-SiC(0001)), or sapphire (Al_2O_3(0001)). The crystalline quality of AlN layers was studied as a function of different parameters: quality and type of the initial substrate, growth parameters and growth protocol for different epitaxy techniques such as ammonia (NH_3) molecular beam epitaxy (MBE), plasma-assisted MBE or metal-organic chemical vapor deposition (MOCVD). Ex situ tools such as Atomic Force Microscopy...
(AFM), Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM), X-ray diffraction (XRD) and in situ Reflection High-Energy Electron Diffraction (RHEED) were used to characterize and evaluate the quality of the layers. However, if the resolution of these tools is high enough to determine the density of microscopic defects, they are limited to determine precisely the dislocation density exhibited by these layers. STM was used to study GaN layers (3.4 eV gap) and proved to have sufficiently high resolution to reveal the terminations at the surface of screw (SD) and edge (ED) dislocations. But the large gap of AlN (6.2 eV) forbids the use of STM on this surface. Introduced by Albrecht et al. in 1991, non contact AFM (NC-AFM) is now well known for its ability to achieve true atomic resolution images on insulator or large gap semiconductor surfaces and thus is a powerful tool to study and characterize the AlN surfaces.

In spite of its 13 % in-plane lattice mismatch with AlN(0001), Al₂O₃(0001) substrate is the most used industrially for growing nitride compounds. However, the growth on Si(111) substrates, which are less expensive, should offer an industrial advantage. But due to its higher lattice mismatch (≈ 19%) with AlN, the growth of AlN(0001) layers with low dislocation density on Si(111) substrates is still challenging. The most relevant substrate to grow low defect AlN layers is nH-SiC(0001) because of its very low in-plane lattice mismatch of 1%, but its cost and the difficulties to prepare its surface make it less used than Al₂O₃.

The goal of this study was to characterize by NC-AFM under UHV the AlN(0001) surfaces grown by NH₃ MBE on two different substrates: Si(111) and 4H-SiC(0001). With the NC-AFM technique we can observe directly two kinds of dislocations (screw and edge) visible at the surfaces as well as the roughness at the atomic scale. By adjusting the growth parameters we show that the dislocation density visible at the surface can be decreased by at least a factor 10, reaching a value of 20/µm² (2×10⁹/cm²) on 4H-SiC(0001) substrates. Ex situ SEM and XRD measurements were also performed to determine the quality of these layers in order to compare them to the state of the art.

II. EXPERIMENTAL

AlN samples are grown by NH₃ MBE in a UHV chamber with a base pressure of 10⁻¹⁰ Torr equipped with a RHEED gun working at 15 keV and a video acquisition system. Al and Si atom beams are provided by high temperature effusion cells and ammonia (NH₃) is used as nitrogen precursor. The substrates are heated radiatively by means of a graphite heater with a maximum temperature of 1300°C. The temperature is measured by an infrared pyrometer, with an estimated incertitude of ± 30°C for silicon and ± 50°C for SiC substrates.

The preparation of the Si(111) substrates begins with the removal of the native oxide by immersion in a 1% solution of hydrofluoric acid (HF). The RCA process is used to grow a thin oxide layer of 10 nm to protect the sample during its mounting on the Omicron plate specific to the NC-AFM microscope. This oxide layer is removed thermally under UHV by annealing the sample at 800°C until the (7 × 7) reconstruction is observed by RHEED. The temperature is then decreased to 600°C and the sample exposed to NH₃ for 10 seconds at 10⁻⁶ Torr followed by annealing at 800°C to promote the formation of crystalline Si₃N₄ areas. A first Al monolayer (ML) is then deposited at 600°C. The temperature is increased to 950°C while maintaining the Al beam and increasing the NH₃ pressure to its growth value around 10⁻⁵ Torr. The growth is continued during the time necessary to get a thickness between 75 nm and 250 nm with a growth rate of 100 nm/h.

This is the typical growth process developed by the CHREA-CNRS laboratory.

The 4H-SiC substrates received from the CREE company are first processed by H₂ gas etching at 1350°C for 30 min at 20 Torr. This process allows the smoothing of the surface and the apparition of large terraces. The sample is then introduced into the MBE chamber and annealed at 1100°C. One to two ML of Si are deposited, leading to the apparition of the (√3 × √3)R30° reconstruction on the RHEED pattern. From there, two different growth protocols were used. The first one follows the same steps as for the Si(111) substrate. In the second one, the initial and final 5 nm of the film are grown at a low growth speed of 10 nm/h by decreasing the Al beam flux, while the central part is grown at 100 nm/h.

After growth, the sample is transferred under UHV to a NC-AFM microscope for surface characterization. The NC-AFM microscope is a commercial room-temperature (RT) ultrahigh vacuum
STM/AFM (Omicron NanoTechnology GmbH, Taunusstein, Germany). QNCHR silicon cantilevers provided by NanoSensors (Neuchatél, Switzerland) were used, with no special preparation except for a moderate heating (150°C) in vacuum. The resonance frequencies were close to 280 kHz, with quality factors ranging from 35,000 to 45,000. The images were obtained in the constant ∆f mode with an oscillation amplitude from 2 to 5 nm. The maximum size of NC-AFM images is 500 nm × 500 nm and the minimum size is several nm². The NC-AFM topography images allow to observed the defects at the atomic and nanometric scale on the atomically flat surface, such as the terminations at the surface of bulk dislocations. To obtain their density, we count them on 3 to 5 NC-AFM images of (500 nm)² area.

After these in-situ NC-AFM experiments, samples were also characterized ex-situ by SEM and some of them by XRD. The XRD measurements were performed on a Panalytical Empyrean device. The used wavelength was produced by a Co anticathode (λ_kα1 = 1.789 Å). The beam monochromatization was obtained by using a hybrid primary optics (mirror + 2-bounce Ge(111)). The samples were mounted on a χ,ϕ,z stage. The secondary optics consisted in a parallel plate collimator in order to achieve a good resolution for the measurements of the (0002) (symmetric set-up) and (10̅13) (asymmetric set-up) reflections.

III. AlN(0001) GROWN ON Si(111) SUBSTRATES

Typical NC-AFM topography images of Si(111)-(7×7) and AlN(0001)/Si(111) surfaces are shown in Fig. 1(a) and 1(b) respectively. The Si(111) surface presents large terraces separated by 1.6 Å high monoatomic steps. It exhibits white dots, 0.5 to 5 nm high, with a density around 240/µm². The origin of these nanodots is related to the impurities present in the chemical solutions used during the surface preparation.

The step edges that appear on the AlN(0001) surface (Fig. 1(b)) are 2.5 Å high, corresponding nicely to c/2 where c=5.185 Å is the lattice parameter of AlN in the direction perpendicular to the (0001) surface. They are aligned along the [10̅10], [01̅10] and [1̅120] directions (<10̅10> family directions). As observed on GaN(0001) by STM, the step-edge intersections form 60° angles, leading to a typical triangular shape morphology. The AlN surface exhibits two families of defects: large hexagonal hillocks visible on the SEM image of Fig. 2(b) and terminations of dislocations at the surface. Note that in the following we will refer to the terminations at the surface of edge and screw dislocations as “edge dislocation” (ED) and ”screw dislocation” (SD). The edge dislocations and screw dislocations appear as small depressions or “pits” in the surface visible as black dots in the NC-AFM images. The edge dislocation pointed by the black arrow in the NC-AFM image of Fig. 1(b) is detailed in Fig. 1(c): the cross section shows a depth of 3 Å for the “pit”. The screw

![FIG. 1. AlN(0001) grown on Si(111) : (a) - NC-AFM image of the Si(111)-(7×7) surface obtained after annealing at 800°C under UHV. The insert shows the (7×7) reconstruction. (b) - NC-AFM image of a typical AlN(0001) surface obtained on Si(111). The surface exhibits two different kinds of dislocations: screw dislocations and edge dislocations pointed by the white and black arrows. The height of the AlN steps is 2.5 Å. (c) - Details of an edge dislocation : the cross section along the black arrow shows a depth of 3 Å. (d) - Details of a pair of screw dislocations. Image parameters: (a) - 500x500 nm², amplitude=2 nm, ∆f=-10 Hz, insert: 11×11 nm², amplitude=2 nm, ∆f=-25 Hz; (b): 500x500 nm², amplitude=2 nm, ∆f=-10 Hz.]
FIG. 2. AlN(0001) grown on Si(111): (a) - RHEED pattern (15 KeV) along the [11\overline{2}0] axis of a 10 nm thick AlN layer at the beginning of the growth. (b,c) - SEM images of 100 nm (b) and 250 nm (c) thick AlN layers. The 100 nm thick AlN layer exhibit 50 nm wide hexagonal defects with a density of $3 \times 10^7$ defects/cm$^2$. The 250 nm thick AlN layer relaxed by cracking along the $<10\overline{1}0>$ crystallographic directions.

The NC-AFM observations of the EDs and SDs on AlN(0001) surfaces are similar to what was observed by STM on GaN(0001) surfaces.\textsuperscript{11} The strength of the NC-AFM technique is to directly observe the dislocations on the AlN(0001) surfaces without the need for specific preparation like the KOH etching which is usually used to reveal the dislocations before SEM imaging. The atomic resolution of the NC-AFM technique is high enough to ensure that all the dislocations that terminate at the surface are indeed observed.

On the surface of this AlN layer grown on Si(111), the average dislocation density is $\approx 600/\mu m^2$ ($\approx 400$ SDs/$\mu m^2 + \approx 200$ EDs/$\mu m^2$). With such a high density, the largest area without dislocation visible at the surface is typical of the emergence of mixed or pure dislocations with Burgers vector of $\frac{1}{3}(11\overline{2}3)$ type or (0001) type respectively. Since we are not able to distinguish between them at the surface, the term screw type will therefore refer either to pure screw or mixed dislocations. The screw dislocations appear always by pair with opposite Burgers vectors as shown in the NC-AFM images of Fig. 1(d): The step around the left SD turns clockwise, while the step around the right SD turns anticlockwise.
dislocations has a maximum size of $(50 \text{ nm})^{2}$. To better understand the origin of these dislocations, we studied the impact on the dislocation density of the quality of the Si(111) surface, the growth temperature, the thickness and the III/V ratio.

The preparation of the Si(111) $(7 \times 7)$ reconstructed surfaces leads to surfaces with different nanodots density. The Si(111) surface shown in Fig. 1(a) is an example of the best surface obtained with a nanodot density $\approx 240/\mu\text{m}^{2}$ and terraces 100 to 200 nm wide. AlN layers grown on Si(111) surfaces including up to five times more nanodots and smaller terraces (10 to 50 nm wide) show no significant increase of the dislocation density, which stays around $700/\mu\text{m}^{2}$. This clearly highlights that the nanodots are not the main seeds for the creation of bulk dislocations in AlN(0001) grown on Si(111).

Growth temperatures from 900°C to 1100°C were studied. It is known that the temperature influences the diffusion length of atoms adsorbed on the surface and then the size of terraces. However it did not affect the dislocation density. Excessive temperature (T\text{substrate} > T\text{Al sublimation} $\approx 1,100^\circ\text{C}$) significantly increases the roughness by creating one monolayer high holes induced by Al atoms sublimation.

AlN film thicknesses from 75 nm to 250 nm were studied. Because the AlN film is initially very rough and mostly 3D as seen on the RHEED pattern for a 10 nm thick AlN layer (see Fig. 2(a)), it is not possible to perform NC-AFM characterization below 50 nm. From 75 to 200 nm, the dislocation density stays constant. After 200 nm the surface relaxes by cracking along the $<10\bar{1}0>$ directions as shown in Fig. 2(c) for a 250 nm thick layer. However, no clear decrease of the dislocation density was observed after the relaxation of the AlN layer.

We arbitrarily defined the III/V ratio $R_{\text{III/V}} = 1$ for $P_{\text{NH}_3} = 5.10^{-6}$ Torr and a beam equivalent pressure for Al (BEP$_{\text{Al}}$) of $3.10^{-8}$ Torr. $R_{\text{III/V}}$ was adjusted from 0.5 to 12 by increasing or decreasing $P_{\text{NH}_3}$ or BEP$_{\text{Al}}$. The study was performed on one sample, first grown with $R_{\text{III/V}} = 1$ for a thickness of 150 nm. The sample was transferred into the MBE chamber after each NC-AFM characterization and $\approx 5$ nm thick layers were added with $R_{\text{III/V}}$ increased from 0.5 to 12. For this range of III/V ratio, the roughness of the surface remains similar, contrary to what is observed in MOVPE growth. However an effect is noticed on dislocation densities: the SD density decreases from $\approx 250/\mu\text{m}^{2}$ to $\approx 180/\mu\text{m}^{2}$ after the second growth at $R_{\text{III/V}} = 3$. It remains constant around $160-180/\mu\text{m}^{2}$ for the other values of $R_{\text{III/V}}$. In contrast, the ED density increases significantly when $R_{\text{III/V}} = 12$, where it reaches twice the values observed for lower $R_{\text{III/V}}$. Table I presents the dislocation densities vs the ratio $R_{\text{III/V}}$.

This study shows that it is not possible to decrease significantly the SD density for AlN layers grown on Si(111). Nevertheless, the hole density can be slightly decreased by lowering the temperature during growth and the $R_{\text{III/V}}$ at the end of the growth. The best quality obtained shows a SD density around $160/\mu\text{m}^{2}$ and an ED density around $250/\mu\text{m}^{2}$ for the optimized growth protocol, i.e. growth rate $= 100$ nm/h, $P_{\text{NH}_3} = 10^{-5}$ Torr, BEP$_{\text{Al}} = 3.10^{-8}$ Torr, temperature $= 900^\circ\text{C}$ and thickness $\approx 160$ nm. However, the SD density remains important and leads to atomically flat terraces without dislocations smaller than $(50 \text{ nm})^{2}$. The XRD measurements made on this best layer gives FWHM values of 1250 arcsec and 970 arcsec for the (0002) and (1013) peaks respectively, while for the worst layer they reach the values of 2050 and 1450 arcsec. The lower values are comparable to the ones obtained by Moreno et al. at the CRHEA laboratory.

| $P_{\text{NH}_3}$ $10^{-6}$ Torr | BEP$_{\text{Al}}$ $10^{-8}$ Torr | $R_{\text{III/V}}$ | SD density $/\mu\text{m}^{2}$ | ED density $/\mu\text{m}^{2}$ | Total $/\mu\text{m}^{2}$ |
|-------------------------------|-------------------------------|-----------------|----------------|----------------|----------------|
| 3 | 10 | 0.5 | $160 \pm 40$ | $460 \pm 60$ | $620$ |
| 3 | 5 | 1 | $248 \pm 40$ | $568 \pm 60$ | $816$ |
| 3 | 1.7 | 3 | $184 \pm 60$ | $536 \pm 60$ | $720$ |
| 6 | 1.7 | 6 | $148 \pm 60$ | $456 \pm 60$ | $604$ |
| 6 | 0.8 | 12 | $152 \pm 75$ | $1044 \pm 80$ | $1196$ |
IV. AlN(0001) GROWN ON 4H–SiC(0001) SUBSTRATES

An \( \text{H}_2 \) gas etching process \((1,350^\circ\text{C}, 20 \text{ Torr}, 30 \text{ min})\) is applied ex situ to the SiC(0001) substrate before the growth of AlN, in order to get a surface with large terraces \((2 \text{ to } 3 \mu\text{m})\) separated by 4 monolayers high steps. The residual defects \((\approx 3/\mu\text{m}^2)\) are eliminated after the introduction of the sample in the MBE chamber by annealing at \(\approx 1100^\circ\text{C}\) under a \(\text{Si}\) flux of \(0.1 \text{ ML/s}\). Depending on the quantity of \(\text{Si}\) deposited, the surfaces can present coexisting reconstructions: \((\sqrt{3} \times \sqrt{3} \ R30^\circ)\), \((12 \times 12)\), \((6 \times 6)\), \((4 \times 8)\), \((14 \times 14)\) and \((3 \times 3)\). In the NC-AFM image of Fig. 3(a), a different reconstruction was identified on each terrace, thanks to atomic resolution studies. But if the \(\text{Si}\) beam flux is too high and the substrate temperature too low, the SiC(0001) surface is less homogeneus with the apparition of \(\text{Si}\) clusters and surface reconstruction domains of smaller area. In the NC-AFM of Fig. 3(e) the dark holes have a height of around 1Å and the \(\text{Si}\) clusters appear as white dots pointed by the dark arrow. As shown in the following, the homogeneity of the prepared SiC(0001) surfaces has a huge effect on the crystalline quality of the AlN layers.

AlN sample A (Fig. 3(b), 3(c), 3(d)) was grown on the \(\text{SiC}\) surface of Fig. 3(a) and sample B (Fig. 3(f)) on the \(\text{SiC}\) surface of Fig. 3(e). The main difference compared to AlN layers grown on Si(111) is the absence of edge dislocations: only pairs of screw dislocations are visible on the NC-AFM images and hexagonal hillocks on the SEM images. The maximum dislocation density is \(\approx 150/\mu\text{m}^2\), 2.5 times lower than for the best AlN layer grown on Si(111). The presence on the \(\text{SiC}\) surfaces of \(\text{Si}\) clusters and of step edges has a direct influence on the hillocks density as observed on the SEM images: The less the surface is homogeneous, the more the hillocks density is raised. XRD measurements confirms this trend. For sample A the FWHM values for the (0002) and (1013) peaks are 370 and 610 arcsec, while they reach 1050 and 900 arcsec for sample B. By using an atomically flat \((\sqrt{3} \times \sqrt{3} \ R30^\circ)\) reconstructed 4H-SiC(0001) surface, we succeeded in reducing by a factor \(\approx 2\) the SD density \((\approx 80/\mu\text{m}^2)\) and to 0 the hillock density. In this case the FWHM values for the (0002) and (1013) peaks decrease to 150 and 430 arcsec respectively.

Compared to our best AlN layer grown on Si(111), these FWHM values are divided by a factor

![Fig. 3](image-url)

**FIG. 3.** 200 nm thick AlN grown on 4H-SiC(0001): (a) - NC-AFM image of a 4H-SiC(0001) surface after 5 cycles of \(\text{Si}\) deposition. The steps are 1 Å high. (b) - NC-AFM image of a 200 nm AlN thick layer grown on the 4H-SiC(0001) sample shown in (a). (c) - RHEED pattern \((15 \text{ KeV})\) of sample A along the \([10\overline{1}0]\) axis after growth of the first 5 nm. (d) - SEM image of the 200 nm AlN layer shown in (b). (e) - NC-AFM image of a 4H-SiC(0001) surface after 10 cycles of \(\text{Si}\) deposition. The dark arrow points on a \(\text{Si}\) cluster. (f) - SEM image of the AlN layer obtained on the 4H-SiC sample shown in (e). Image parameters: (a) -500x500 nm\(^2\), amplitude=5 nm, \(\Delta f=-40 \text{ Hz}\); (b) -500x500 nm\(^2\), amplitude=5 nm, \(\Delta f=-20 \text{ Hz}\); (d) -11x8 \(\mu\text{m}^2\); (e) -500x500 nm\(^2\), amplitude=5 nm, \(\Delta f=-10 \text{ Hz}\); (f) -19x14 \(\mu\text{m}^2\).
FIG. 4. 5 nm thick AlN grown on 4H-SiC(0001): NC-AFM image of the surface of 5 nm thick AlN(0001) layer grown on 4H-SiC(0001). The surface exhibits edge dislocations which are located at the interface between the crystalline domains A and B rotated by 180° as shown by the triangular shape of the islands outlined with dashed lines. The dark lines indicate the border between the domains. Image parameters: 500×500 nm², amplitude=5 nm, Δf=−5 Hz.

8 and 2 for the (0002) and (10 ¯13) peaks, confirming the better quality of the AlN layer grown on 4H-SiC(0001).

Thicknesses from 5 nm to 400 nm were studied. According to the RHEED pattern, the beginning of the growth at 100 nm/h is 3D, and becomes 2D after the growth of 25 to 30 MLs. There is no significant reduction of the SD density between 200 nm and 400 nm grown in the same condition. However, for a very low growth rate (10 nm/h) of a 5 nm thin AlN layer, it is possible to reduce the SD density to \( \approx 0/\mu m^2 \). The sharp lines, the intensity and the Kikuchi lines of the RHEED pattern displayed in the Fig. 3(c) for such a layer indicate that the surface is extremely smooth and flat, which is confirmed by the NC-AFM image shown in Fig. 4. This surface presents EDs visible on the NC-AFM images and also on the SEM images in Fig. 5. Their origin will be discussed in the next section. Nevertheless, the areas between these EDs are atomically flat with a size that reaches (200 nm)².

V. ORIGIN OF DEFECTS

Thanks to these two studies with high resolution NC-AFM images, it was possible to show that the first MLs of the growth have a direct influence on the hillock and the dislocation densities. For AlN grown on Si(111), the main origin of the dislocations is the in-plane lattice mismatch of 19 % between AlN(0001) and Si(111). This mismatch generates a high dislocation density located at the interface between the AlN layer and the Si substrate and may lead to an initial 3D growth. These dislocations propagate through the layer and terminate at the surface in the form of edge dislocations and pairs of screw dislocations. A thickness of about 50 nm is necessary to bury the initial 3D nucleation layer before reaching a 2D growth regime. After optimizing the growth parameters and the preparation of the Si(111)-(7 × 7) reconstructed surface, the minimum SD and ED densities induced by the lattice mismatch are about 200/\(\mu m^2\) (2×10¹⁰/cm²).

For a 5 nm AlN layer grown on 4H-SiC(0001), the NC-AFM image of Fig. 4 shows that the EDs are mainly placed on the borders between crystallographic domains rotated by 180°, as shown by the orientation of the triangular islands on the same surface monolayer. These borders are usually called stacking mismatch boundaries (SMBs). They are induced by the polytype mismatch between 4H-SiC and 2H-AlN. If the diffusion of species is efficient enough to allow the nucleation at the 4H-SiC step edges, the EDs linked to the SMBs should be close to the step edges. On the
SEM images of Fig. 5, lines of EDs pointed by white arrows are visible. Their orientation and their density are typical of the orientation and the step edge density of the initial 4H-SiC substrate. Besides these lines of EDs, there are aggregation of EDs pointed by dark arrows. We have observed after the growth of 200 nm of AlN a density of hillocks that is similar to the density of these ED aggregations, suggesting that the latter are the seeds for the former. Besides the presence of hillocks on the 200 nm thick AlN layer, no more EDs are observed on the NC-AFM images, but only pair of SDs. This indicates that the density of crystallographic domains growing up to the surface of the film is drastically reduced: locally, one domain expands at the expense of the other during the growth, leading to buried interfaces between the two types of domains, that are likely to generate threading dislocations appearing as pairs of SDs at the surface of the 200 nm thick layer. But compared to AlN layers grown on Si(111), the total SD density is always lower for 200 nm thick AlN layer, up to 10 times at ≈ 20/μm² (2×10⁹/cm²).

VI. AlN(0001) GROWN AT 10 nm/h

The effect of a low growth rate of 10 nm/h on the surface roughness and on the dislocation density was studied by comparing two samples with thickness around 200 nm. For the first one the growth was realized with the usual protocol at 100 nm/h during 2 hours followed by 30 min at 10 nm/h. For the second one a starting layer was realized with a growth rate of 10 nm/h during 30 min and then the same growth sequence as for the first sample. As expected, the end of the growth at 10 nm/h reduces the overall roughness of the surface (fig 6(b) and 6(d)) with island and hole densities decreased by at least a factor of five compared to the surface obtained at 100 nm/h presented in figure 3(b). Such smooth surfaces with atomically flat terraces of several hundred of
square nanometers are suitable for the growth of thin quantum well and the formation of quantum dots induced by thickness fluctuations as already observed on Al$_{1-x}$Ga$_x$As/GaAs quantum wells.$^{18}$

The main effect of a low growth rate is observed on the dislocation density, when the growth is started at 10 nm/h during 30 min. In these conditions, the SD density is around 20/µm$^2$ (fig 6(d)), five times lower than for films started at 100 nm/h (fig 6(b)). This improvement of the layer quality can be explained by the diminution of the nucleation density during the first monolayer growth. This effect is observed on the RHEED patterns after the growth of 0.9 ML: While the pattern obtained for a growth rate of 100 nm/h (fig 6(a)) shows a 3D behavior, the one at 10 nm/h (fig 6(c)) shows a perfect 2D layer. The low growth rate of 10 nm/h allows to begin the 2D growth after the first ML while more than 25 MLs are necessary for that at 100 nm/h.

For 10 nm thick layers grown at 10 nm/h, the influence of $R_{III/V}$ equal to 1/30 or to 1/10 was also studied by increasing or decreasing the NH$_3$ pressure while the Al beam flux remained constant. The SD density stayed between 0 and ≃ 10/µm$^2$ and the general aspect of the surface was not significantly affected.

VII. SUMMARY

The use of NC-AFM coupled under UHV to a MBE chamber dedicated to the growth of AlN layers by ammonia molecular beam epitaxy has offered a unique way to characterize the surfaces of the Si(111) or the 4H-SiC(0001) substrates and the surfaces of the AlN layers grown on them in different conditions. The terminations of threading dislocations and their density can be rapidly determined and correlated to the surface conditions of the substrate and the growth parameters. Coupling these in-situ images to ex-situ SEM images allowed us to show the origin of threading dislocations in AlN(0001). For AlN layers grown on Si(111), the remaining nanodots on the Si(111) surface do not seem to have an impact on the dislocation density and therefore dislocations are mainly induced by the in-plane lattice mismatch of 19% between AlN(0001) and Si(111). By optimizing the preparation of the Si(111) surface and the growth parameters we obtained AlN layers with a terminating dislocation density of $2 \times 10^{10}$/cm$^2$ and FWHM values for the XRD (0002) and (10 13) peaks of 1250 and 970 arcsec respectively. For 200 nm thick AlN layers grown on 4H-SiC(0001), no edge dislocations (ED) are visible at the surface. The 4H-SiC surface conditions impact mainly the hillock density generated by the aggregation of edge dislocations that appear at the beginning of the growth (5 nm). Others EDs related to the stacking fault boundaries (SMBs)
are aligned mainly along the step edges of the 4H-SiC surface. They are buried after 200 nm growth where only SD dislocations are visible at the surface. By decreasing the growth speed from 100 nm/h to 10 nm/h at the beginning of the growth and continuing with the normal growth speed of 100 nm/h, the terminating dislocation density can be decreased to its lower value of $2 \times 10^9$ cm$^{-2}$ and the FWHM values for the XRD (0002) and (1013) peaks are also decreased to 150 and 430 arcsec respectively.

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