Journal Name RSCPublishing

ARTICLE

Silane controlled three dimensional GaN growth and recovery stages on cone-shape nanoscale patterned sapphire substrate by MOCVD

Cite this: DOI: 10.1039/x0xx00000x

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Received 00th January 2012,
Accepted 00th January 2012

DOI: 10.1039/x0xx00000x

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Three dimensional (3D) growth induced by silane was performed on the cone-shape nano-scale patterned sapphire substrates (NPSS) by metalorganic chemical vapor deposition (MOCVD). The growth evolution for silane controlled 3D growth process and the recovery stage were investigated by a series of growth interruptions. The GaN epilayers grown on the templates with different 3D growth conditions were characterized by X-ray diffraction (XRD), Raman scattering, and atomic force microscopy (AFM) measurements. The full width at half maximums (FWHMs) of (002) and (102) reflections in XRD rocking curves were 267 and 324 arcsec, respectively for the sample on NPSS with 600 s 3D growth. And the extremely smooth surface with the average roughness was achieved as 0.10 nm scaled in $3\times3\mu m^2$. All the above data were superior to those for the planar sample or NPSS ones without optimized 3D growth time. The silane addition caused the effective 3D growth. The proper size and homogeneity, faceted sidewall of islands by 3D growth led to high crystalline quality, much strain relaxation and specular surface of GaN epilayers.

Introduction

Patterned sapphire substrate (PSS), which is orientated from the epitaxially lateral overgrowth (ELOG) technique[1], has become a popular commercial technology for GaN-based light emitting diodes (LEDs). Both crystalline quality and light extraction efficiency (LEE) of LEDs can be improved significantly[2, 3]. A trend of the PSS technology is to shrink the pattern size to get more light extraction[4 - 7] and to resolve the wafer bowing problems in GaN growth on large size sapphire substrate (more than 4 inch)[8, 9] and in thick film growth, such as 300 μm or more on heterogeneous substrate[10]. Nano PSS (NPSS) has been paid much attention on the fabrication of high efficiency LEDs in recent years. Almost all of the reports agree with that the LEE for NPSS is superior to that for conventional PSS. However the crystalline quality improvement is still in dispute for different groups.

Ashby *et al*. predicted nm-scale features would be necessary to reduce more threading dislocation density (TDD) than that for μm-scale one[1]. Some researchers believe that the TDD could be reduced by compliance “bridges” from the mismatched epilayers to the nano-patterned substrates[11-14]. The low angle grain boundary defects can be healed at the nano heteroepitaxy[14]. It is reported that the TDD of GaN epilayer on NPSS is reduced 2 orders by the abbreviated nucleation process[9]. The bending or blocking of the TDs is observed near the interface of NPSS/GaN epilayer[4]. Su *et al*. reported that the crystalline quality of GaN on NPSS was inferior to that on conventional μm-scale PSS, since the coalescence fronts in the defective region was more close to the interface[5].

The mechanism of GaN initial growth stages on micro-PSS has been reported by some groups[15 16]. However, there are few reports about initial growth process for NPSS. It is found that the surface energy of sapphire substrate has been altered by nano patterns[9]. It is well known that the surface energy affects the absorption, diffusion and desorption of the adatoms on the growth surface. The surface reaction and atoms arrangement on the crystal surface may be also influenced by the nano patterns. Silicon atoms are used as a “surfactant” for GaN quantum dots growth on AlGaN surface[17]. Pakula *et al.* reported that the TDD in herteroepitaxial GaN was reduced about 2 orders by 300 s silane treatment[18]. The vertical growth rate enhancement with increasing silane flow rate has also been reported in GaN-based nanorods growth[19]. Silicon acted as a surfactant is seldom used in GaN growth on NPSS, which may delay the nucleation island coalescence to reduce the TDD[20] or inhibit the TD propagation upward[18]. Liu *et al*. also reported the stress at the interface between GaN and AlN template delayed the island coalescence and led to high quality GaN growth[21].

In the work, the abbreviated nucleation process and two-step undoped-GaN (u-GaN) growth was used similarly to our previous work[22]. The difference was that the silane was introduced during three dimensional (3D) GaN growth stage after buffer deposition and annealing, which substituted the conventional one by low V/III ratios. The 3D growth evolutions were studied by a series of growth interruptions. Their effects on the crystalline quality, surface morphology and strain relaxation were also studied on the coalesced samples. The roles of silane on the 3D GaN growth were concluded.

Experiment

Two-inch c-plane sapphire substrates were used to fabricate the cone-shape NPSS by nano-imprint lithography (NIL) using Obducat Eitre3 instrument, and inductively coupled-plasma (ICP) dry etching using Samco RIE-200ip etcher. The pitch and the size of the hexagonal arrangement pattern were 550 and 420 nm, respectively. The height of the pattern was 245 nm. After the patterning process, the growth of GaN was carried out by using low pressure metal organic chemical vapor deposition (MOCVD) in the Thomas Swan close coupled showerhead reactor. GaN growth were carried out by using trimethylgallium (TMGa) and ammonia (NH_3) gases as the source materials for Ga and N, respectively. Hydrogen (H_2) was used as the carrier gas. Prior to the GaN growth, the substrates were cleaned in piranha (H_2SO_4:H_2O_2 = 3:1) and 15% HCl solution, and then loaded into MOCVD reactor. GaN epilayers were grown in this sequence, a low-temperature (530℃) GaN buffer layer was first grown with the reduced time of 150 s[22]. Afterwards, the chamber temperature was raised to 1020℃ for the recrystallization of GaN buffer layers, followed by the two-step high temperature growth. The first step was carried out with 1 sccm silane flow rate and the second one was the recovery stage in ordinary lateral growth conditions. The conditions for the two steps were same except the silane flow rate. For the silane contributes to 3D growth as the antisurfactant[17], the lateral growth was found to be suppressed in the first stage. So this stage was denoted as 3D growth in this work. Meanwhile, GaN layer directly grown on the planar sapphire and GaN grown on NPSS without silane were used as the reference samples. Both steps were performed and interrupted for observing the GaN 3D islands growth and coalescence.

The crystalline quality of the GaN epilayers grown on both NPSS and planar sapphire substrate was studied by X-ray diffraction (XRD: Bruker D8, Cu Kα1 X-ray source λ = 1.5406Å) rocking curves. Micro-Raman scattering experiments were performed on the above samples by a Renishaw Confocal Raman microscope with a 514.5 nm laser and 1 cm^{-1} resolution. The samples are characterized by atomic force microscopy (AFM: Bruker Dimension Icon) to study the surface morphology. In addition, the scanning electron microscopy (SEM: Nova Nano SEM 430) were performed to record the islands growth and coalescence processes.

Results and discussion

Fig. 1 shows the results of XRD rocking curves of GaN epilayers grown on planar sapphire and NPSS. The GaN epilayers were grown on NPSS without and with 3D growth for 300, 600 and 800 s, respectively, as shown in Fig. 1a. After the

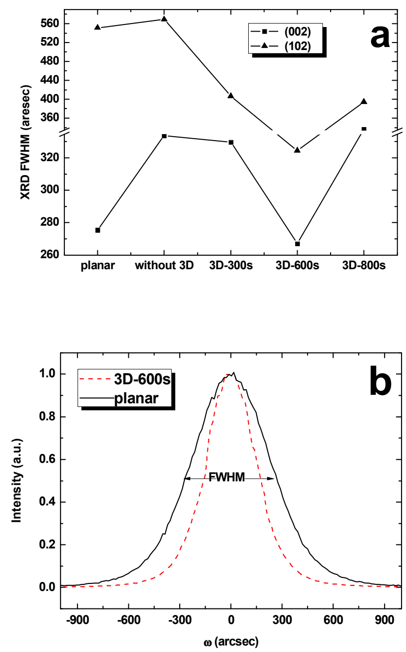

Fig.1 (a) the dependence of XRD FWHM of GaN films on different 3D growth time for on planar sapphire and NPSS. (b) XRD rocking curves of (102) plane for planar samples and NPSS with 3D growth time for 600 s.

3D growth, all the samples were grown in recovery stage for 2400 s. It is observed that without 3D growth, the full width at half maximum (FWHM) values for (002) and (102) planes are higher than those for other samples. With the 3D growth time increasing, FWHMs for (002) and (102) planes are decreased till 600 s 3D growth, and then increases. The smallest FWHMs for (002) and (102) planes are 267 and 324 arcsec for 3D-600s sample, while those for planar samples are 275 and 551 arcsec, respectively. The significant difference of FWHMs for (102) plane is shown in Fig. 1b between the planar and NPSS 3D-600s samples. It is well known that the (002) FWHM can demonstrate screw TDD, and (102) FWHM corresponds to screw and edge TDD. The screw and edge TDs are usually generated by tilt and twist of the columnar grain boundaries, respectively. Without 3D growth, the GaN island coalesced thickness is as low as 100 nm on AGOG NPSS while 250 nm for planar one[9]. According to in-situ laser reflectance curves for the growth processes (not shown here), the reflectivity increases and begins to vibrate earliest for the without 3D sample, which means GaN islands coalesce to smooth rapidly. It is not suitable for the early coalescence to obtain high crystalline quality with low TDD, which is conformed to the previous report to delay the islands coalescence[20]. Certainly, the excessive 3D growth also degrades the crystalline quality when the growth time is higher than 600 s.

To further compare the residual stress in GaN films on planar sapphire and NPSS with different 3D growth time, the Raman spectra were measured and are shown in Fig.2. There are two phonon modes are Raman active, i.e. E_2 (high) and A_1 (LO) modes

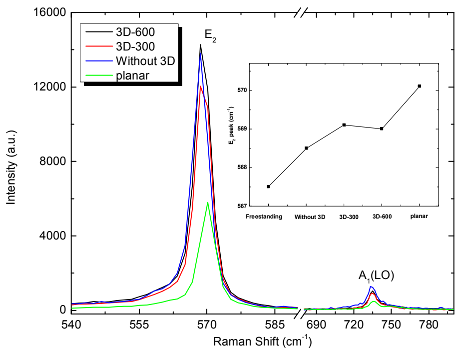

Fig.2 Raman spectra of GaN films grown on planar sapphire and NPSS with different 3D growth time. The inset is E_2 peaks for different GaN films fitting by Gaussian function, the start point is for the freestanding GaN which is free of strain.

in each spectrum. E_2 peaks for NPSS samples show obvious red-shift compared to that for the planar one. The A_1 (LO) peaks keep constant for the all of the four samples. NPSS without 3D growth shows the smallest E_2 peak as 568.5 cm^{-1}, while 3D-600 and 3D-300 show 569.0 and 569.1 cm^{-1}, respectively. The E_2 peak for planar sample is about 570.2 cm^{-1}. And E_2 peak for a 400 μm thick free-standing GaN estimated as strain free is also measured as 567.5cm^{-1}. The E_2 peaks for different GaN films are shown in the inset in Fig.2. The E_2 (high) mode phonon scattering was used to characterize the in-plane compressive stress distribution using the relation $\Delta\omega=K\sigma_x$. Here pressure coefficient K=4.3 cm^{-1} GPa^{-1}, $\Delta\omega$ is the phonon peak shift, and σ_x is the biaxial stress. The compressive stresses in GaN films are obtained as 0.23, 0.34 and 0.63 GPa for NPSS without 3D growth, with 600 s 3D growth and planar samples, respectively. Sample without 3D shows less in-plane compressive stress in the next recovery growth film. With 3D growth, more compressive stress is introduced in the recovery stages. However, all three NPSS samples show less strained than the planar one. The strain relaxation for NPSS samples can be attributed to more lateral overgrowth than that for planar one. Without 3D growth, the recovery stage shows more lateral overgrowth because the GaN islands are smaller and the recovery centre density are more than those with 3D growth. Smaller uniform islands can also coalescence to form the smooth surface earlier, but it also causes larger TDD in the films, as indicated in XRD curves. For 3D-600 sample, there is a bit more strain relaxation than 3D-300 sample because of the Ostwald ripening and crystal facet formation, as described later.

The surface morphologies of GaN films grown on planar sapphire substrate and the NPSS with and without 3D growth were further characterized by AFM, as shown in Fig.3. The root mean square (RMS) for GaN film grown on planar sapphire substrate is 0.35nm, while those for GaN on NPSS without and with 3D growth stage are 0.22 and 0.10 nm, respectively. The smooth surface will benefit to the full LED structure growth in sequence. It is clearly observed from well-arranged steps and terraces that the lateral growth mode is dominant. The AFM image shows a more highly parallel ordering of atomic steps on the surface of the layer grown on NPSS with a 600 s 3D growth. As to the planar sample, the parallel steps and terraces can be observed too. But the spacing of the steps is not uniform. Although without 3D growth sample shows the smoother

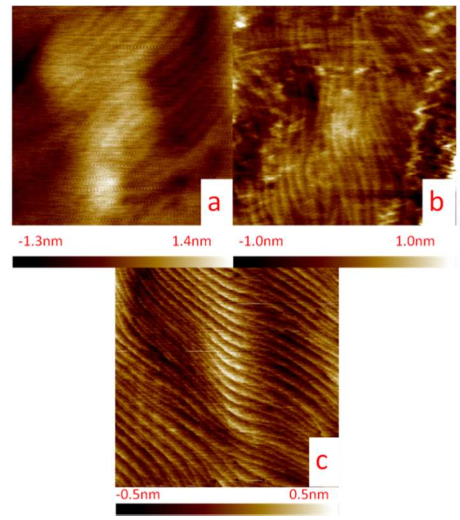

Fig. 3 AFM images of GaN films grown on (a) the planar sapphire substrate (b) the NPSS without 3D growth and (c) the NPSS with 3D growth. All topographic AFM images are scaled in 3μm×3μm.

morphology than that for planar one, the steps and terraces are bunched and disordered. The extremely smooth morphology is achieved on GaN surface with 3D growth for 600 s, which is also smoother than the samples for longer 3D growth time. The sample without 3D growth was still smoother than the planar sample. It can be explained that smaller and more uniform GaN islands on NPSS would lead to a smooth coalescence. The coalescence will affect the final roughness of the GaN films. However, the earlier coalescence will produce more defects, which will disturb the step flow growth and show the disordered the steps. The detailed process for 3D growth and its recovery will be further discussed in the following SEM measurements.

The first high temperature step for 3D growth may play a significant role of controlling the recovery center density and developing crystalline facets. Both of them are important to the lateral growth stage[16]. The 3D growth evolution studies of the GaN epitaxy on NPSS in silane atmosphere were carried out. Firstly, the SEM images of cone- shape NPSS are shown in Fig. 4a and b in bird view and side view, respectively. The pitch and size of the pattern are 550 and 420 nm, and the height is about 245 nm. Fig. 4c-f reveal the 3D growth behaviors of GaN islands with silane for 100, 300, 600 and 800 s, respectively. The initial 3D growth occurs at the interval between the patterns, which seems quite uniform due to the ordered NPSS. There is no wetting phenomenon of GaN layer on the NPSS since several contrast spaces are not filled up.

With the 3D growth going on, GaN islands grow up and start to coalesce in the intervals gradually. If there is no silane added, the coalescence will be completed rapidly, according to Ref. 9 and our growth reflectance curves. Moreover, the speed of the top surface area increasing is lower than that of the height increasing, especially for the isolated GaN islands. It indicates that the lateral growth rate is less than the vertical one in the 3D growth stage. The silane should be acted as the antisurfactant, by which the growth surface energy on NPSS is changed, and the diffusion length of adatoms may be reduced. The silane controlled 3D growth is more convenient and lower cost than other methods, such as low V/III ratio or low temperatures.

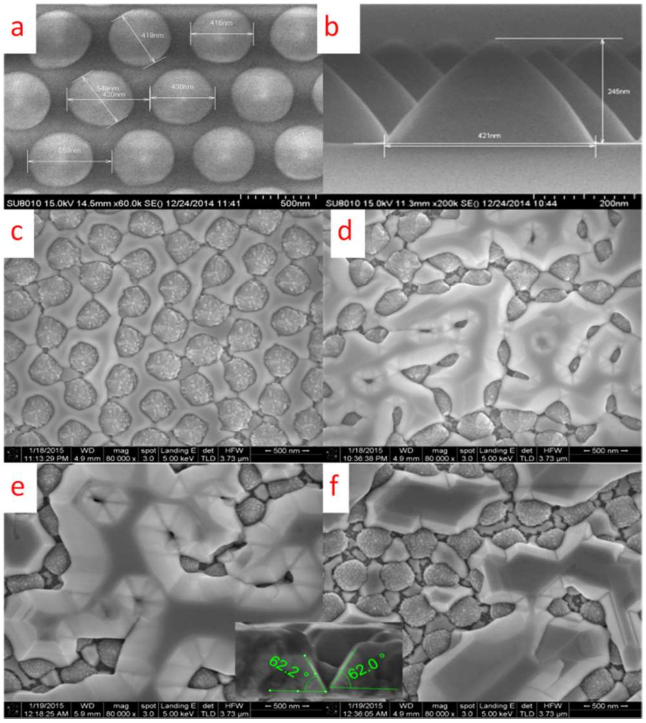

Fig. 4 SEM images of the cone-shape NPSS with (a) bird view and (b) side view, and GaN 3D growth behaviors for (c) 100 (d) 300 (e) 600 and (f) 800 s, respectively. The inset shows the cross-sectional SEM image of the typical pattern for (e) and (f)

There are two important phenomenon happened in 3D growth stage. One is that the sidewall of these GaN islands are formed by $\{1\bar{1}01\}$ facets, which is tilted with an angle of 62° to the c-plane of the substrate, as shown in the inset of Fig. 4e and f. As sample with 300 s 3D growth, the crystalline facets are not clearly observed. The crystalline facets are appeared when they grown most slowly. The pattern acts as a growth mask which will change the surface energy for lower height GaN islands. Formation of these facets has also been observed during selective GaN growth on stripe/dot patterned sapphire[23,24]. Those GaN facets would promote the suppression of dislocation by introducing the image force in lateral growth stage. Therefore it is observed in XRD rocking curves that the FWHMs of (002) and (102) are smaller for with 3D growth samples than those for without 3D growth samples. FWHMs for 3D-600 sample are less than those for 3D-300 sample. The other phenomenon shows that some islands in the interval of the patterns disappear with the enlargement of the other GaN islands in the intervals. This interesting phenomenon can be explained by the well-known Ostwald ripening mechanism [25]. The Ostwald process would introduce the size inhomogeneity of the GaN islands, which would reduce the recovery center density and increase the difficulty of smooth coalescence in a short growth period. Fig.6f shows the larger GaN islands and more digested spaces between the patterns for 800 s 3D growth, which experiences a bit longer Ostwald process. According to the XRD results, too long 3D growth time is hard to provide a uniform surrounding for GaN islands lateral growth in the recovery stage.

It is notable that the size of GaN islands by 3D growth on NPSS is much less but more homogenous than those for planar one and μm-scale PSS[16]. The strain is relaxed more for NPSS than that on planar one. Moreover, the larger size and inhomogeneity of the GaN islands also lead to larger roughness. Certainly, the recovery growth process is another important factor affecting the surface roughness, as described in the following.

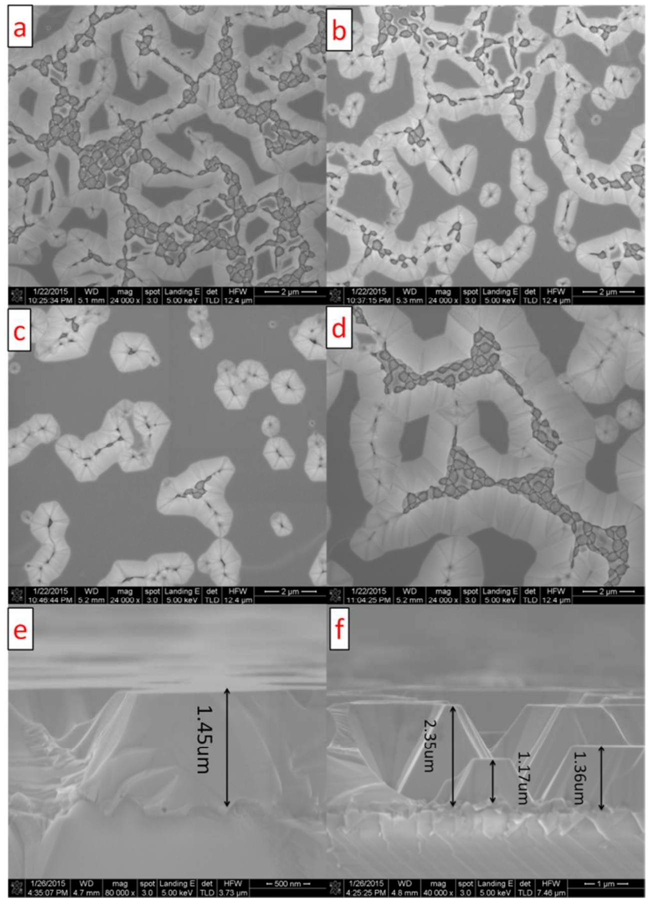

Fig.5 SEM images of GaN recovery growth for a) 300, b) 600, c) 1200 s on the templates with 600s 3D growth, and d) 1200s on the template with 800 s 3D growth model; (e) and (f) were cross-sectional images of 1200s on the template with 600 and 800 s 3D growth stage.

The recovery stages were also investigated in different lateral growth time on the same 3D growth templates and same lateral growth time on the different 3D growth templates. SEM images of the GaN recovery growth for 300, 600 and 1200 s on the optimized templates with 600 s 3D growth are shown in Fig. 5a-c. Compared with the Fig.4e, GaN islands with flat top surface grow up and recover the nano-pattern gradually with the growth time increasing. The size and height distribution of the GaN islands are rather uniform. Combing the bird view and side view images as shown in Fig.5c and e, the GaN islands coalescence is almost finished with only few patterns seen after 1200s lateral growth. It is indicated that coalescence GaN film with smooth surface can be obtained on NPSS within a very short growth period. The growth time would be shortened more by further reducing the size of the NPSS pattern and optimizing the lateral growth conditions.

In order to verify the optimization of the 3D growth, sample of 1200 s GaN recovery growth on the template with 800 s 3D growth was also studied, as shown in Fig.5d and f. Compared with the Fig.4f, some islands grow up and the nano-patterns nearby are recovered while the rest areas maintain the original unfilled nano-pattern, as shown in Fig.5d. The Fig.5f displays the very large difference of the height for the islands nearby with 1200s recovery growth on the template with 800 s 3D growth. Three typical GaN islands are measured as 1.17, 1.36 and 2.35 μm, respectively.

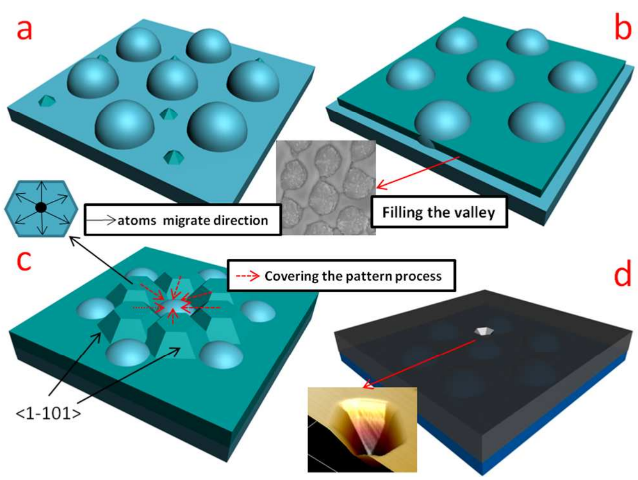

Fig. 6 Schematic drawing of the two-step growth mode procedure of GaN epitaxy on the NPSS. (a) Buffer layer and nucleation, (b) 3D growth, (c) 3D growth and recovery growth, (d) recovery growth.

However, the slant angles of all the sidewall of the GaN islands keep 62^{o} to the c-plane of substrate. The morphology observation is agreed with the previous XRD and AFM results. As a deduction, the larger and inhomogeneous of GaN islands affects the recovery growth significantly. Some GaN islands enlarge more based on the larger cluster formed in 3D growth stage. Simultaneously, some areas remain intact for lacking recovery centers, which disappeared in the 3D growth stage. Such disordered growth surrounding to the recovery centers leads to a long lateral recovery period and thereby the correspondingly rough morphology eventually.

To explain the evolution of initial GaN growth on the NPSS more vividly, a simplified sketch is provided in Fig. 6. After the low-temperature buffer layer growth followed by a temperature ramping process to 1020 ℃, GaN islands grow in the intervals, as shown in the Fig. 6a. Owing to the nm-scaled pattern, the GaN islands keep small size and distribute uniformly. Followed, the GaN islands almost fill up the space between the nano patterns swept over the valley firstly although the silane controlled 3D growth causes the GaN layer not well wetting, as shown in Fig. 6b. The islands grow up from the bottom of the NPSS with r-plane $\{1\bar{1}01\}$ formed on the sidewalls, as shown in the Fig. 6c. The directions of the GaN islands covering the pattern and the movements of atoms on the top surface of the GaN islands are drawn in the Fig.6c. Finally, the GaN islands coalescence with smooth surface is completed and the inverted hexagonal pyramid pits are formed, diminished and disappeared eventually as shown in Fig.6d. Due to the uniformity of the GaN islands and faceted sidewall, the strain relaxed film with smooth surface, low TDDs can be obtained within quite short lateral growth periods.

Conclusions

In summary, MOCVD growths of GaN epilayers have been performed on NPSS with and without silane in 3D growth stage, and on planar sapphire substrate. The silane controlled 3D growth process and its effects on the recovery stage were investigated by a series of growth interruptions. The extremely smooth surface with the average roughness was achieved as 0.10 nm scaled in 3×3 μm^2 for the sample with 600 s 3D growth and 2400s lateral growth on NPSS. The silane addition causes the effective 3D growth. The proper size and homogeneity, faceted sidewall of islands by 3D growth lead to high crystalline quality, much strain relaxation and specular surface of GaN epilayers. It is indicated that the excellent properties could be obtained for 2.4 μm thick GaN films, which is about half that of conventional PSS one. The growth time could be shortened more by further reducing the size of pattern and optimizing the 3D growth conditions.

Acknowledgements

This work was supported by National Key Basic Research Program of China under Grants Nos. 2011CB301905, 2013CB328705, National Natural Science Foundation of China under Grants Nos. 61334009, 60976009, U0834001, and Guangdong Innovative Research Team Program (No. 2009010044).

Notes and references

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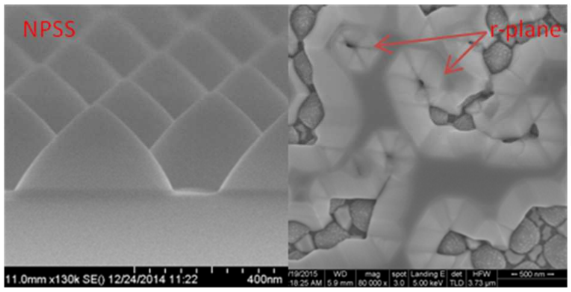

The silane-controlled 3D growth led to high crystalline quality, much strain relaxation and specular surface of GaN epilayers on NPSS.