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Defects and nucleation of GaN layers on (0001) sapphire

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Abstract

The morphology and microstructural evolution of a nucleation layer are analysed using high-resolution transmission electron microscopy. Low-temperature nucleation of GaN on (0001) sapphire is investigated. Depositions were made for 20, 40, 60, 120 and 180 s at 560 °C by metal–organic chemical vapour deposition. It is shown that the shortest deposition times give rise to the formation of cubic islands. Subsequently, the density and the size of the nucleated islands increase and they start to transform into wurtzite from the interface with the substrate. From the start, the nuclei contain misfit dislocations. At these early growth stages, the relaxation state changes from one island to another; this probably underlies the subsequent mosaïc growth of the high-temperature-active GaN layers.

1. Introduction

For the last decade, GaN and related compounds have attracted great interest due to the development of applications such as high-brightness light-emitting diodes [1] and laser diodes [2] operating in the blue/green-to-ultraviolet range. Significant progress has been achieved in the growth process for GaN. Thin films are mostly grown by metal–organic chemical vapour deposition on *c*-plane sapphire, despite its large lattice parameters and thermal coefficient mismatch. The introduction of a low-temperature buffer layer has considerably improved both the morphology and the properties of subsequent high-temperature-grown GaN layers [3]. Much work has been devoted to the factors that can affect the crystalline quality of the buffer layer and therefore the growth behaviour of the high-temperature GaN, such as the growth conditions (temperature, trimethylgallium (TMGa) and NH_3 flux), the thickness and

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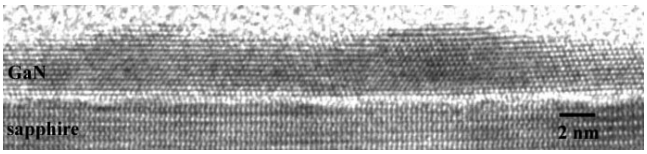

Figure 1. A HREM image of the GaN nucleation layer after 20 s of growth.

the substrate treatment [4]. The purpose of this work is to achieve a better understanding of the growth mechanisms of the GaN nucleation layer and the formation of associated defects.

2. Experimental details

GaN nucleation layers were grown on *c*-plane sapphire substrates by metal–organic chemical vapour deposition. The substrates were cleaned with solvents and subjected to *in situ* pretreatment under a H_2 flow at 1100 °C. The nucleation layers were grown at 560 °C using TMGa and ammonia (NH_3). The investigation growth times were 20, 40, 60, 120 and 180 s. High-resolution electron microscopy (HREM) was used to determine the epitaxy and microstructure of the GaN layers. Specimens were prepared by conventional dimpling and ion milling techniques. Electron microscopy observations were carried out using a Topcon electron microscope operating at 200 kV with a point-to-point resolution of 0.18 nm.

3. Results and discussion

Cross-sectional observations on the 20 s GaN layer were performed first. Cubic islands with the epitaxy $(111)_{GaN} \parallel (0001)_{sapphire}$ and a surface ripple morphology were found and seem to have grown on a sapphire step, as shown in figure 1. This appearance of surface ripple morphology has already been observed in Si_xGe_{1-x} films and is generally interpreted in terms of the Asaro–Tiller–Grinfeld instability [5]. In a previous report [6], we showed by cross-sectional and plan-view TEM analysis respectively the absence of a wetting layer next to the islands and the presence of GaN crystallites of small sizes, which is indicative of a Volmer–Weber growth mode. By using Fourier filtering, misfit dislocations were revealed. The large distance between them means that the islands are highly strained at this very early stage of growth.

When the growth time increases to 40 s, cross-sectional HREM images show isolated and relatively well-spaced 3D islands faceted following the {111} planes, without any wetting layer at the substrate, which confirm the Volmer–Weber growth mode. In a previous study, plan-view observations have shown that the GaN crystallites were triangular or hexagon shaped [6]. Figure 3 illustrates the case of a 40 s GaN faulted island with a hexagonal structure at the interface and cubic areas developing with stacking faults towards the top of the island. Fourier filtering has been performed at the interface on figure 3; it revealed that the island is relaxed via the presence of seven misfit interfacial dislocations (figure 4). Dislocations appear every 6–7 $\{10\bar{1}0\}$ GaN lattice fringes facing every 7–8 $\{11\bar{2}0\}$ Al_2O_3 lattice fringes.

At 60 s growth, islands with different sizes, shapes (somewhat rounded or dome-like) and microstructures are observed and some of them begin to coalesce (figure 5). The coalescence really takes place from 120 s of growth, the shape of islands becoming progressively pyramidal. Hexagonal stacking, stacking faults and microtwins are developing through the islands; the cubic phase is still dominating the microstructure of these islands. Figure 6 is a HREM image of such a specimen. At 180 s of growth, the coalescence has proceeded further and pyramidal islands cover the substrate surface. Cubic and hexagonal areas in the vicinity of the

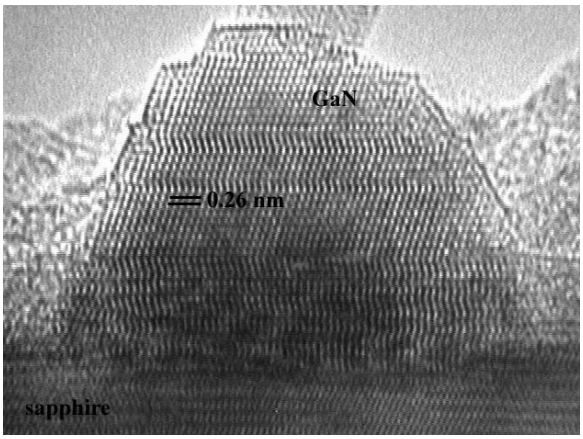

Figure 2. HREM observation of a GaN island after 40 s of growth.

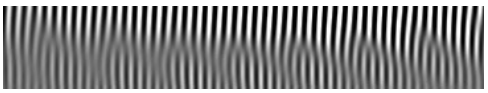

Figure 3. A Fourier-filtered image revealing seven misfit interfacial dislocations, using $\{10\bar{1}0\}$ GaN and $\{11\bar{2}0\}$ Al_2O_3 spots.

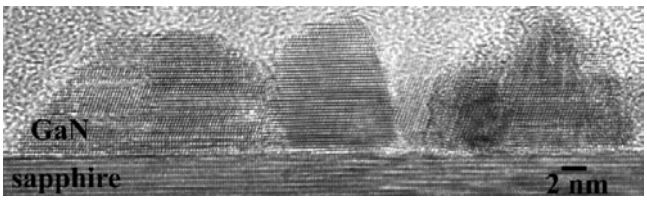

Figure 4. A HREM image showing the coalescence between dome-like GaN islands after 60 s of growth.

Figure 5. A HREM image of the 120 s nucleation layer.

interface and through the islands are larger (the cubic phase still predominates), leading to a higher density of stacking faults and microtwins, as can be seen in figure 7. A grain boundary between two GaN islands is located on the left-hand side of figure 7 (arrow). A rotational symmetry of 180° is also observed between them. This symmetry across the boundary is very often considered as a factor which can lead to the formation of inversion domain boundaries in epitaxial GaN layers [7]. 60° edge dislocations are visualized under the boundary between the two GaN islands but also in the island situated in the right-hand part of the figure.

To estimate the relaxation in GaN islands, calculations of residual strains have been made using the following formula: $\delta = \delta_e + \varepsilon_r$, where δ is the mismatch between GaN and sapphire,

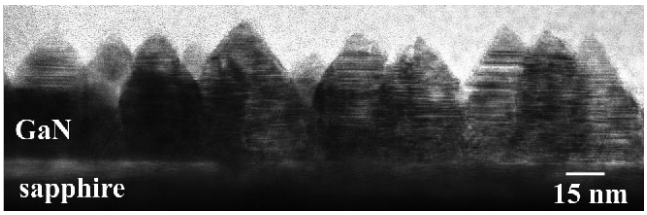

Figure 6. A HREM image of the 180 s nucleation layer showing pyramid-shaped islands.

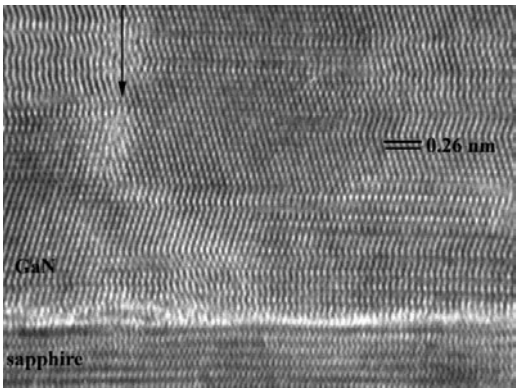

Figure 7. A HREM image taken in the vicinity of the interface and showing the coalescence of two islands at 180 s of growth.

Table 1. Relaxation strain for GaN islands deposited at different growth times.

s	d_e(GaN) (nm)	δ_e	ε_r
20	0.4062	−0.0583	−0.1193
40	0.2749	−0.1555	−0.0054
60	0.2765	−0.1622	+0.0013
180	0.2769	−0.1639	+0.0030

δ_e is the effective misfit in a relaxed configuration and ε_r is the residual strain;

$$\delta = \frac{d_s\{11\bar{2}0\} - d_{\mathrm{GaN}}\{10\bar{1}0\}}{d_s\{11\bar{2}0\}} = -0.1609.$$

d_s and d_{GaN} are, respectively, the corresponding hexagonal lattice parameters of sapphire and GaN, with $d_s\{11\bar{2}0\} = 0.2379$ nm and $d_{\mathrm{GaN}}\{10\bar{1}0\} = 0.2762$ nm. Table 1 gives the average experimental d_e-spacing of GaN, the effective misfit and the residual strain degree for GaN islands at different growth times. For the GaN island deposited at 20 s growth time, $d_s\{11\bar{2}4\} = 1.919$ and $d_{\mathrm{GaN}}\{100\} = 4.52$ Å, corresponding to the hexagonal lattice parameter of sapphire and cubic lattice parameter of GaN, respectively, have been used. The misfit in this case is −0.1777.

We have measured the residual strains using all the misfit dislocations on many islands for each sample. Table 1 gives the results for 20, 40, 60 and 180 s. The large residual strain value obtained at 20 s shows that the GaN island is partially relaxed. Mean values over some 5–10 islands for the following growth times are given. The variations are small and one may be tempted to say that there is a relaxation rate of 100%. In fact, our measurements show that

islands are never equally relaxed which means that when they coalesce their rotations will be different. This is certainly one of the origins of the huge numbers of threading dislocations which are found in the thick GaN layers.

4. Conclusions

Nucleation of GaN on (0001) sapphire gives rise to the formation of islands following the Volmer–Weber growth mode. Coalescence seems to occur from a growth time of 60 s. From the analysis of misfit dislocations, it is found that all the islands tend to reach a maximum relaxation state but this is unequally attained.

Acknowledgments

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References

[1] Nakamura S, Senoh M, Nagahama S, Iwasa N, Yamaha Y, Matsushita T, Kiyoku H and Sugimoto Y 1996 *Japan. J. Appl. Phys.* 2 **53** L74
[2] Nakamura S, Senoh M, Nagahama S, Iwasa N, Yamaha Y, Matsushita T, Sugimoto Y and Kiyoku N 1997 *Appl. Phys. Lett.* **70** 1417
[3] Amano H, Sawaki N, Akasaki I and Toyoda Y 1986 *Appl. Phys. Lett.* **48** 353
[4] Briot O, Alexis J P, Tchounkeu M and Olombard R L 1997 *Mater. Sci. Eng.* B **43** 147
[5] Jesson D E, Chen K M and Pennycook S J 1996 *MRS Bull.* **21** 31
[6] Degave F, Ruterana P, Nouet G, Je J H and Kim C C 2002 *Mater. Sci. Eng.* B **93** 177
[7] Cheng L, Zhou K, Zhang Z, Zhang G, Yang Z and Tong Y 1999 *Appl. Phys. Lett.* **74** 661