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Structural evolution of GaN layers grown on (0 0 0 1) sapphire by hydride vapor phase epitaxy

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ABSTRACT

GaN was grown directly on (0 0 0 1) sapphire by a two-step process using hydride vapor phase epitaxy (HVPE). Nucleation layers deposited on sapphire at ~450–500 °C consisted of localized epitaxial wurtzite GaN nano-crystals. In between and above the epitaxial nano-crystals were randomly oriented wurtzite GaN nano-crystals. GaN islands of various sizes and shapes were formed, after annealing between ~900 and 1000 °C, through a decomposition–redeposition process. Preferential growth of GaN occurred on the islands that had an epitaxial relationship with sapphire during the subsequent high-temperature overgrowth. Threading dislocations were observed in isolated GaN islands that were formed after annealing.

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1. Introduction

Hydride vapor phase epitaxy (HVPE) is currently widely used to grow thick GaN layers on sapphire, mainly due to its very fast growth rate that ranges from tens to hundreds of microns per hour [1–5]. The growth of very thick GaN layers using HVPE, however, usually requires GaN template layers grown by metal–organic chemical vapor deposition (MOCVD) [6–8]. Without such template layers, GaN layers grown directly on sapphire may crack when the thickness of GaN exceeds certain values [9]. It would be more cost-effective and more efficient if we could avoid such MOCVD template layers and grow GaN directly on sapphire without cracking.

There have been some attempts to grow GaN directly on sapphire [9–13] using a growth process that resembles the standard two-step growth process used in MOCVD. Thick GaN layers with acceptable quality can be achieved. For example, Martin et al. [12] reported a threading dislocation density as low as $10^8/\mathrm{cm}^2$ at a thickness of 8 μm, and Tavernier et al. [9] reported a threading dislocation density of $6\times10^7/\mathrm{cm}^2$ at a thickness of 23 μm. Structurally, after high temperature annealing, GaN islands were formed with their distribution resembling GaN islands grown by MOCVD [11,13]. However, there is a lack of knowledge on the structures of the GaN nucleation layers before annealing. It was mentioned that such nucleation layers are nanocrystalline [13]. However, unlike the MOCVD-grown GaN nucleation layers that have been thoroughly studied, no details can be found regarding the structures of the nanocrystalline GaN nucleation layers grown by HVPE. Part of the reason why such layers were overlooked might be due to the fact that empirically the quality of thick GaN layers seems to be rather insensitive to small variations in the low temperature nucleation conditions [13]. This is very different from the situation in MOCVD, where the quality of GaN depends sensitively on both nucleation temperature and nucleation time.

We report here detailed analyses on the structures of GaN nucleation layers grown at low temperatures using HVPE. The structural evolution that GaN underwent during the subsequent annealing and the early stage of overgrowth will also be reported. Based on this knowledge, we are able to explain why the quality of HVPE-grown GaN layers does not depend sensitively on the nucleation conditions.

2. Experimental details

The GaN layers were grown directly on (0 0 0 1) sapphire using a proprietary hydride vapor phase epitaxy (HVPE) reactor. $GaCl_3$ and NH_3 were used as sources and a N_2/H_2 mixture served as the carrier gas. We performed a full factorial design of engineering experiments in order to optimize growth conditions, including nucleation temperature and time as well as annealing temperature and time. In this paper we discuss samples that are representative in terms of microstructure, yet not necessarily the best templates in terms of threading dislocation density in thick GaN layers grown on them.

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Table 1 lists the growth conditions of 7 samples presented here. The 7 samples include one nucleation layer, two annealed layers and a sequential growth of four samples that include one annealed layer and three interrupted overgrowth layers with time intervals of 15 s.

An atomic force microscope (AFM) was used for surface morphology scans. Both cross-sectional and plan-view microstructural analyses on these samples were performed on a JEOL 4000EX TEM operating at 400 kV.

3. Results

Fig. 1(a)–(d) shows cross-sectional TEM images taken from sample #1, a GaN nucleation layer deposited on (0 0 0 1) sapphire at 500 °C for 18 s. Fig. 1(a) shows a low magnification image taken along the $\langle 1\,\bar{1}\,0\,0\rangle$ direction of sapphire. A white arrow in this image indicates the interface between the GaN nucleation layer and sapphire. The nucleation layer is continuous, around 40 nm thick, with surface roughness at a nanometer level. Fig. 1(b) shows a selective area diffraction pattern (SADP) taken along the $\langle 1\,\bar{1}\,0\,0\rangle$ direction of sapphire from a region contiguous to the GaN/sapphire interface. The large, round and bright spots in the pattern are due to single crystalline sapphire. Besides these spots, there exist diffraction rings passing through diffraction spots that are tiny and weak. These tiny and weak spots are due to wurtzite GaN that grew epitaxially on sapphire, in the orientation of GaN [0 0 0 1]$\parallel$sapphire [0 0 0 1] and GaN $[1\,\bar{1}\,0\,0]\parallel$sapphire $[1\,1\,\bar{2}\,0]$, as revealed by the pattern. The correlation between the diffraction rings and the diffraction spots from GaN indicates that there are randomly oriented wurtzite GaN crystals in the nucleation layer. The diffraction ring in the center is due to GaN (0 0 0 2). The other diffraction ring that is hardly visible is due to GaN $(1\,\bar{1}\,0\,3)$. Both diffraction rings are indicated by dashed arrows.

Table 1
Growth conditions of 7 samples.

Run #	**Nucleation**		**Annealing**		**Overgrowth**	
	Temp. (°C)	Time (s)	Temp. (°C)	Time (min)	Temp. (°C)	Time (s)
1	500	18				
2	500	18	1000	1		
3	450	48	900	2		
Sequential growth						
4	475	24	950	1		
5	475	24	950	1	925	15
6	475	24	950	1	925	30
7	475	24	950	1	925	45

Fig. 1(c) shows a cross-sectional high-resolution electron microscopy (HREM) image taken along the $\langle 1\,\bar{1}\,0\,0\rangle$ direction of sapphire, from the GaN/sapphire interface of sample #1. A few GaN nano-crystals that are less than 10 nm in size can be observed. All the nano-crystals in view have wurtzite structure based on their "**AaBbAaBb**" type of stacking sequence. The stacking sequence in one of them is illustrated by two straight lines passing through "**Aa**" and "**Bb**" positions. Some of the GaN nano-crystals are epitaxial with sapphire, while others are not, as indicated in the region marked by the dashed circle. The epitaxial nano-crystals contributed to the tiny and weak GaN diffraction spots shown in Fig. 1(b). However, the GaN grown subsequently above the epitaxial nano-crystals are randomly oriented and embedded in an amorphous-like matrix. Fig. 1(d) shows a cross-sectional HREM image taken along the $\langle 1\,\bar{1}\,0\,0\rangle$ direction of sapphire from the surface of sample #1. Again, randomly oriented nano-crystals embedded in an amorphous-like matrix can be observed. In some of the nano-crystals, the [0 0 0 1] direction of GaN can still be identified and is indicated by the white arrows. Such randomly oriented GaN nano-crystals contributed to the diffraction rings revealed in Fig. 1(b).

Fig. 1(e) shows a plan-view HREM image taken along the [0 0 0 1] direction of sapphire from sample #1. This image was taken near the edge of a hole in the plan-view TEM specimen. A straight dashed line was drawn to roughly mark the interface between the TEM specimen and a thin amorphous layer that was

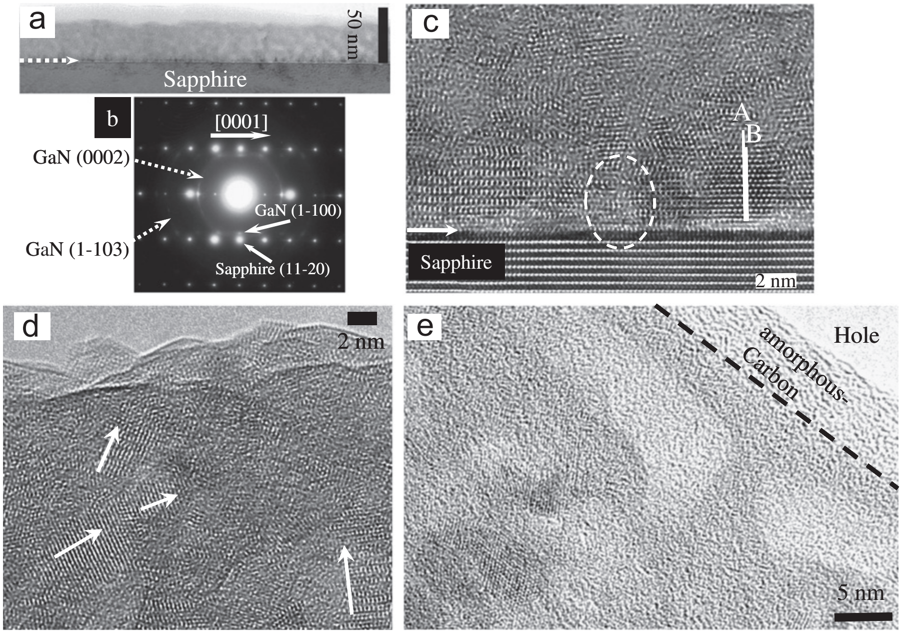

Fig. 1. (a) Cross-sectional bright-field TEM image and (b) an SADP, both taken along the $\langle 1\ -1\ 0\ 0\rangle$ direction of sapphire, from sample #1. The white arrow in (a) indicate the GaN/sapphire interface. (c) Cross-sectional HREM image taken along the $\langle 1\ -1\ 0\ 0\rangle$ direction of sapphire from the GaN/sapphire interface of sample #1. The dashed circle marks an area with non-epitaxial GaN. Two straight lines indicate the stacking sequence in one of the nano-crystals. (d) Cross-sectional HREM image taken along the $\langle 1\ -1\ 0\ 0\rangle$ direction of sapphire from the surface of sample #1. The white arrows indicate [0 0 0 1] direction in GaN nano-crystals. (e) Plan-view HREM image taken along [0 0 0 1] of sapphire, from sample #1. The dashed line marks the boundary between the TEM sample and carbon contamination during TEM observation.