Low threading dislocation density in GaN films grown on patterned sapphire substrates

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Abstract: The growth process of three-dimensional growth mode (3D) switching to two-dimensional growth mode (2D) is investigated when GaN films are grown on cone-shaped patterned sapphire substrates by metal-organic chemical vapor deposition. The growth condition of the 3D–2D growth process is optimized to reduce the threading dislocation density (TDD). It is found that the condition of the 3D layer is critical. The 3D layer keeps growing under the conditions of low V/III ratio, low temperature, and high pressure until its thickness is comparable to the height of the cone-shaped patterns. Then the 3D layer surrounds the cone-shaped patterns and has inclined side facets and a top (0001) plane. In the following 2D-growth process, inclined side facets coalesce quickly and the interaction of TDs with the side facets causes the TDs to bend over. As a result, the TDD of GaN films can decrease to 1×10^8 cm^{-2}, giving full-width at half maximum values of 211 and 219 arcsec for (002) and (102) omega scans, respectively.

Key words: threading dislocation; GaN; pattern sapphire substrate; metal organic chemical vapor deposition
DOI: 10.1088/1674-4926/33/11/113002 **EEACC:** 2520

1. Introduction

III-nitride material is called the third generation of semiconductors in which tremendous progress has been achieved in recent decades. Due to the remarkable advantages such as wide band gap, chemical and physical stability, and special electrical and optical properties, III-nitride material is widely applied to fabricate light emitting diodes (LEDs), laser diodes (LDs), and high electron mobility transistors (HEMTs)[1, 2]. Generally, sapphire is commonly used as the substrate for III-nitride LEDs. However, the large lattice mismatch (16%) and thermal expansion coefficient mismatch between GaN and sapphire substrate cause high threading dislocation density (TDD) in the range of 10^8–10^{10} cm^{-2}[3]. TDs are very harmful to electronic and optoelectronic devices because they act as non-radiative electron–hole recombination centers[4]. Thus, it is always desirable to reduce the number of TDs in GaN.

There have been several different growth approaches taken to reduce the TDD in GaN films recently. Epitaxial lateral overgrowth (ELOG)[5] and its derivatives can significantly reduce TDD, but it inevitably interrupts the growth process. Meanwhile, growing on patterned sapphire substrates (PSS) is another alternative way and it can also reduce TDD efficiently[6]. In this study, in order to reduce TDD, a 3D–2D growth process[7] was applied and cone-shaped patterned sapphire substrates were used as substrates. During the 3D growth process, the growth condition was adjusted to control the facets including $\{1\bar{1}01\}$ inclined facets and a top (0001) plane. The thickness of the 3D layer should be close to the height of the cone-shaped patterns on which there was little GaN deposited. At the end of the process, cone-shaped patterns were surrounded by a 3D layer. In the following 2D-growth process, inclined side facets coalesce quickly and the interaction of TDs with the side facets causes the TDs to bend over. Here, we varied the growth time and the pressure of 3D growth process, and use X-ray diffraction (XRD), an atom force microscope (AFM), a transmission electron microscope (TEM) and a scanning electron microscope (SEM) to characterize the GaN layer properties.

2. Experimental procedure

In this study, all the GaN films were grown by using a Thomas Swan close coupled showerhead (CCS) MOCVD where a 633 nm laser interferometer was used to monitor the growth. Trimethyl gallium (TMGa) and ammonia (NH_3) were used as precursors and pure hydrogen was used as the carrier gas. At the onset, the PSSs whose specification was 2 μm (bottom) × 1 μm (space) × 1.5 μm (height) were annealed at 1060 ℃ in H_2 atmosphere, the reactor was cooled to 530 ℃ and an about 30 nm thick GaN nucleation layer was deposited. A 3D layer was grown on the nucleation layer, which recrystallized by ramping up the temperature to 1045 ℃ with the rate of 1.5 ℃/s and holding the temperature for about 90 s. During this period, low temperature (about 1010 ℃) and V/III ratio (about 670) were set to encourage the 3D growth. In addition, in order to optimize the parameters of 3D layer growth, the pressure was varied from 4.5×10^4 Pa (sample A) to 6.5×10^4 Pa (sample C) and the growth time was adjusted from 1000 s (sample A) to 850 s (sample D) and 750 s (sample E). After the 3D layer, a 2D growth process for fast merging was recommended. Then the temperature (1060 ℃) and the V/III ratio

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Received 12 April 2012, revised manuscript received 28 April 2012

Table 1. Growth condition for samples A–F.

Sample	Pressure of 3D layer (10^4 Pa)	Growth time of 3D layer (s)
A	4.5	1000
B	5.5	1000
C	6.5	1000
D	4.5	850
E	4.5	750
F	—	—

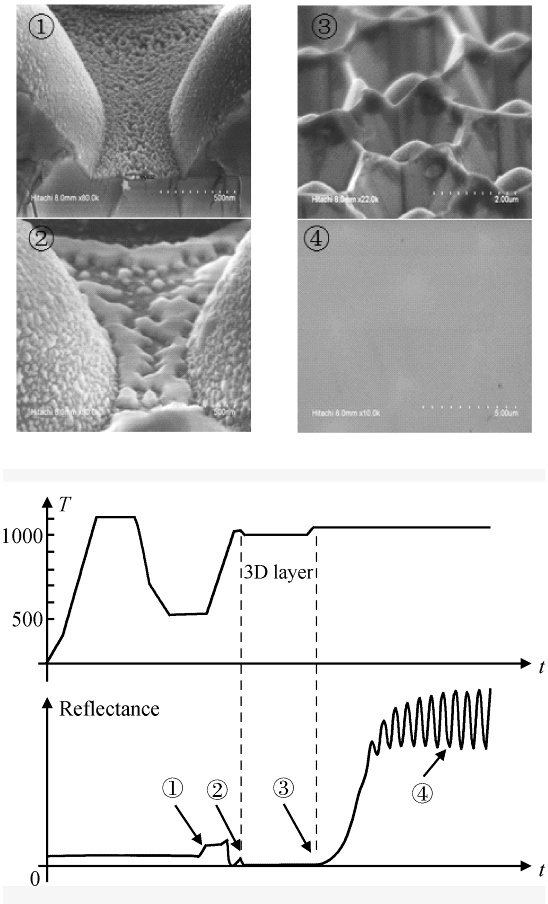

Fig. 1. 3D–2D growth temperature and reflectance transient taken from sample A. SEM insets show the morphology at different stages of growth as the nucleation ① before and ② after recrystallization, ③ at the end of 3D layer and ④ after coalescence.

(about 1200) were increased, and pressure set to 2.0×10^4 Pa. Furthermore, another sample (sample F) was grown by using a normal process for comparison with the 3D–2D process. A summary of the growth parameters for all samples is shown in Table 1.

After growth, morphologies were examined by using an SEM at different stages of sample A. Moreover, TDD was assessed by etch-pit density (EPD) measurements and XRD was used to support the results.

3. Results and discussion

First, we studied on morphological evolution of GaN layer growth with the 3D–2D process. Figure 1 shows the growth temperature and reflectance transient taken from sample A and the morphology at different stages of growth. As shown in inset ①, amorphous GaN is deposited uniformly on the PSS after the nucleation. And the thickness of nucleation layer is about 30 nm. In the duration, the reflectivity increases with the nucleation layer growing because the reflectivity is in proportion to the thickness of nucleation layer. As the nucleation layer recrystallized under a high temperature, the materials are redistributed. GaN grains are formed and then they gain or lose particles from one grain and transport to another, thus smaller grain shrinks while larger grains grow. Finally, platelet-shaped islands are mainly left on the space of PSS, as can be seen in inset ②. In the 3D layer growth, growth conditions are adjusted to ensure that the vertical growth rate is much higher than the lateral growth rate. A 3D layer surrounds the cone-shaped pattern, and the thickness of the 3D layer is near to the height of the cone-shaped pattern. Furthermore, the hexagonal recesses have $\{1\bar{1}01\}$ incline facets and a narrow (0001) plane. In the following 2D process, the lateral growth rate is much higher than the vertical growth rate, thus the incline facets coalesce quickly and the interaction of TDs with the side facets causes the TDs to bend over. However, TDs in the (0001) surface will not bend over. The phenomenon can be observed in Fig. 2, a TEM image of sample E. In Fig. 2, dislocations above the patterns bend over (in circle 1), while those among the patterns extent to the surface (in circle 2).

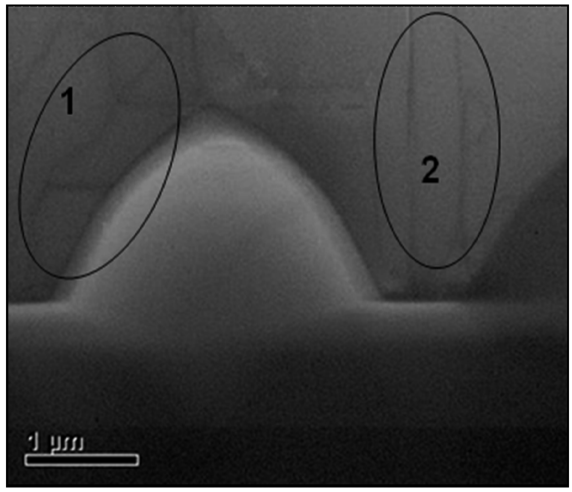

Fig. 2. TEM image of sample E.

The condition of the 3D layer is critical to reduce the TDD. Pressure and growth time are changed for the samples. All samples are etched in hot H_3PO_4/H_2SO_4 (3/1) solution; then EPD is examined by using an AFM. And XRD is used to calculate TDDs. Figure 3 shows the XRD (002) and (102) rocking curves of sample A. Figure 4 shows the AFM image (5 μm $\times$ 5 μm) of sample A after wet chemical etching. For XRD, the measurements of rocking curves of (002) and (102) are carried out. The densities of screw-type dislocation D_S and edge-type dislocation D_E can be estimated from the XRD FWHM values based on the following formulas[8]:

$$D_\mathrm{S} = \frac{\beta_\mathrm{S}^2}{4.35 \times |b_\mathrm{s}|^2} = \frac{\beta_{002}^2}{4.35 \times (b_\mathrm{s} \cos\alpha)^2}, \qquad (1)$$

Table 2. Structural properties of samples.

Sample	FWHM (arcsec)		Dislocation density (10^8 cm^{-2})		EPD (10^8 cm^{-2})
	(002)	(102)	Screw-type	Edge-type	
A	211	219	1.68	0.391	0.98
B	239	242	2.16	0.164	1.36
C	253	288	2.42	2.15	1.64
D	276	287	2.88	0.703	1.92
E	288	315	3.13	1.85	2.52
F	320	425	3.87	8.88	7.00

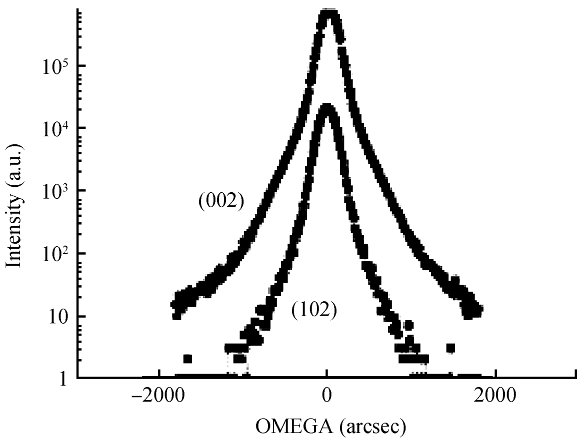

Fig. 3. XRD (002) and (102) rocking curves of sample A.

$$D_{\mathrm{E}} = \frac{\beta_{\mathrm{e}}^2}{4.35 \times |b_{\mathrm{e}}|^2} = \frac{\beta_{102}^2 - \beta_{002}^2}{4.35 \times (b_{\mathrm{e}} \sin\alpha)^2}, \quad (2)$$

where $|b_{\mathrm{s}}|$ and $|b_{\mathrm{e}}|$ are the Burgers vector sizes of the screw-type dislocation ($|b_{\mathrm{s}}| = 0.5185$ nm) and edge-type dislocation ($|b_{\mathrm{e}}| = 0.3189$ nm), respectively, β_{002} and β_{102} are XRD FWHM values of (002) and (102). And α is the angle between the reciprocal lattice vector (K_{hkl}) and the (001) surface normal. By doing this, the calculated results of TDDs are listed in Table 2.

From Table 2, we can see that the TDDs of samples A–E, which were grown using the 3D–2D process, is much lower than that of the sample F, which was grown using the normal process. Besides, TDDs increase as the pressure increases. And TDDs increase as the growth time decreases. Since the pressure also effects on the growth rate, both pressure and growth time relate to the thickness of 3D layer. Thus, the thickness is a critical parameter for reducing the TDDs. The 3D layer has the incline facets $\{1\bar{1}01\}$ and (0001) plane, TDs in the (0001) surface will not bend over, so a small area of (0001) plane is good for reducing TDDs. Generally, the thickness of the 3D layer is recommended to be near to the height of cone-shaped pattern to further reduce TDDs. Normally, the density of screw-type dislocation is lower than that of edge-type dislocation, but the result of the experiment is opposite. Maybe the lateral growth plays an important role, but the exact reason for this needs study further.

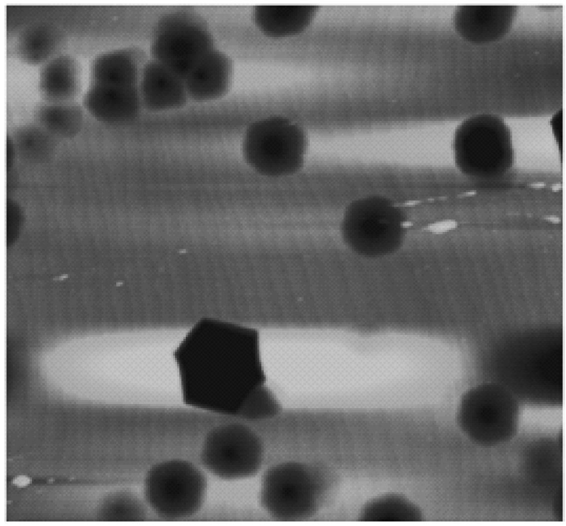

Fig. 4. AFM image (5 μm × 5 μm) of sample A after wet chemical etching.

4. Conclusion

In this paper, we report our research work on the 3D–2D growth process when GaN films grow on cone-shaped patterned sapphire substrates (PSS) by metal-organic chemical vapor deposition (MOCVD) to reduce TDDs. The condition of the 3D layer is critical to reduce the TDDs. During the 3D growth process, we found that TDDs increase as the pressure increases and increase as the growth time decreases. Moreover, the thickness of the 3D layer is recommended to be close to the height of the cone-shaped pattern to further reduce TDDs. In the following 2D-growth process, the lateral growth plays an important role in bending over the dislocations in the GaN epilayer. The results of XRD and AFM prove that GaN film with low TDDs is obtained.

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