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# Growth of N-polar GaN on vicinal sapphire substrate by metal organic chemical vapor deposition

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**Abstract** The growth and properties of N-polar GaN layers by metal organic chemical vapor deposition (MOCVD) were reported. It is found that N-polar GaN grown on normal sapphire substrate shows hexagonal hillock surface morphology. With the misorientation angles increasing from 0.5° to 2.0° toward the a-plane of the sapphire substrate, the number of the hillock becomes less and less and finally the surface becomes flat one on the sapphire substrate with the misorientation angle of 2°. It is also found that the crystalline quality and the strain in the GaN are greatly influenced by the misorientation angle.

**Keywords** GaN; N-polarity; Metal organic chemical vapor deposition

## 1 Introduction

GaN-based III-nitrides are promising materials which have a lot of applications such as solid state lighting, full color display, laser printers, and high density information storage [1–3]. It is known that GaN has hexagonal crystal structure which shows polar nature. This polar nature has great influence on the growth behaviors and electrical and optical properties [4–6]. For example, most of GaN are epitaxially grown on c-sapphire substrate which leads to either metal(Ga)-polarity or N-polarity. Owing to the better stability of Ga-polarity and/or smooth surface, most of devices such as light-emitting diodes, laser diodes, and high electron mobility transistors (HEMTs) are fabricated on Ga-polarity structure. Moreover, N-polarity GaN also shows its advantages. For example, in the case of HEMTs, N-polarity GaN shows low ohmic contact resistances and reduced short channel effect since the polarization direction is opposite to that of Ga-polarity one [7]. More importantly, the enhancement mode transistor can be realized by using the N-polar GaN-based heterostructure [7]. It was also reported that polarization induced hole doping in compositionally graded AlGaN grown on GaN along the N-polarity direction, which can enhance the p-type doping efficiency and thus leads to great application in light emitting devices, particularly at deep ultraviolet region [8]. In addition, both theoretical and experimental studies show that N-polarization emitters show several advantages over conventional emitters for device parameters such as electron overflow, injection efficiency, and laser threshold current density [9–11].

Unfortunately, to grow epitaxy of N-polarity GaN and its device structure were much less studied in comparison with that of Ga-polarity one. This is partially due to that it is quite difficult to grow the pure N-polarity GaN layer on sapphire substrate, the most popular substrate for nitride epitaxy. Thus, hillock growth morphology is usually observed on N-polar GaN which leads to difficultly achieve quantum structures with sharp interface. Therefore, most studies use either C-polarity SiC substrate or N-polarity GaN bulk substrate, which are quite expensive and not suitable for mass production. In this paper, the growth of flat N-polarity GaN layers by using vicinal sapphire substrate was reported.

## 2 Experimental

GaN layers were grown by metal organic chemical vapor deposition (MOCVD, Axtron) system. Normal sapphire and

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those with misorientation angels of 0.5°, 1.0°, and 2.0° were used as substrates. Trimethylgallium (TMGa) and ammonia ($NH_3$) precursors were used for Ga and N sourses, respectively. $H_2$ was used as carrier gas and the reactor pressure was $1.01 \times 10^{-6}$ Pa. The key point to achieve N-polarity GaN samples was using nitridation process, where the sapphire substrate was exposed to the N precursor at 1,100 °C for 2–3 min after thermal annealing. An AlN ultrathin layer with N-polarity is believed to be formed by this nitridation process, which is similar to the case of relative long time sapphire nitridation process in the molecular beam epitaxy (MBE) [12]. Then low temperature GaN buffer layer with a thickness of 30 nm and high temperature GaN epilayer with a thickness of about 2 μm were grown in sequence. The surface morphology of the samples was measured with an optical microscope (OM). Atomic force microscopy (AFM) was also used to give us the surface roughness (root-mean-square value). Crystalline quality of the samples was investigated by X-ray diffraction (XRD), where the ω-rocking curve of both the symmetric and asymmetric scans are measured. Raman measurement was also performed to investigate the stain status in the samples.

## 3 Results and discussion

Figure 1 shows the OM images of GaN samples grown on sapphire substrate with different mis-orientation angles. It is shown that hexagonal hillock surface morphology is observed in GaN films grown on normal sapphire substrate. The size of each hillock is about 150 μm, which is the usual surface for the N-polar GaN layers grown by MOCVD, as reported by several groups [13, 14]. It is believed that the formation of this kind of big hexagonal hillock structure is due to that the N-polarity is not perfect, i.e., some parts or micrograins show Ga-polarity [13, 14]. Since the growth rate of Ga-polarity is a little higher than that of N-polarity, the big hexagonal grains are formed centered by the Ga-polarity grain in nanoscale. It is shown that the number of hexagonal hillock grains becomes less with the mis-orientation angle increasing to 0.5°, where some areas show half or part of hexagonal grains. When the mis-orientation angle increases to 1.0°, the density of grains becomes much smaller while the grain size also becomes smaller. The surface finally changes to flat one and is free of hexagonal grain when the mis-orientation angle increases to 1.0°. Figure 2 shows the surface morphology of GaN layers investigated by AFM, which shows surface features in much small area in comparison with that shown in Fig. 1. Figure 2a shows the morphology of area free of hillock for GaN layer grown on 0.5°-off sapphire. The surface in these areas is quite flat with rms roughness of as small as 0.6 nm in 10 μm × 10 μm scanned area. This value decreases to 0.35 nm in 3 μm × 3 μm scanned area. It is interesting that there is no step, which is different from the step-flow growth of Ga-polar GaN [15]. This shows advantages to

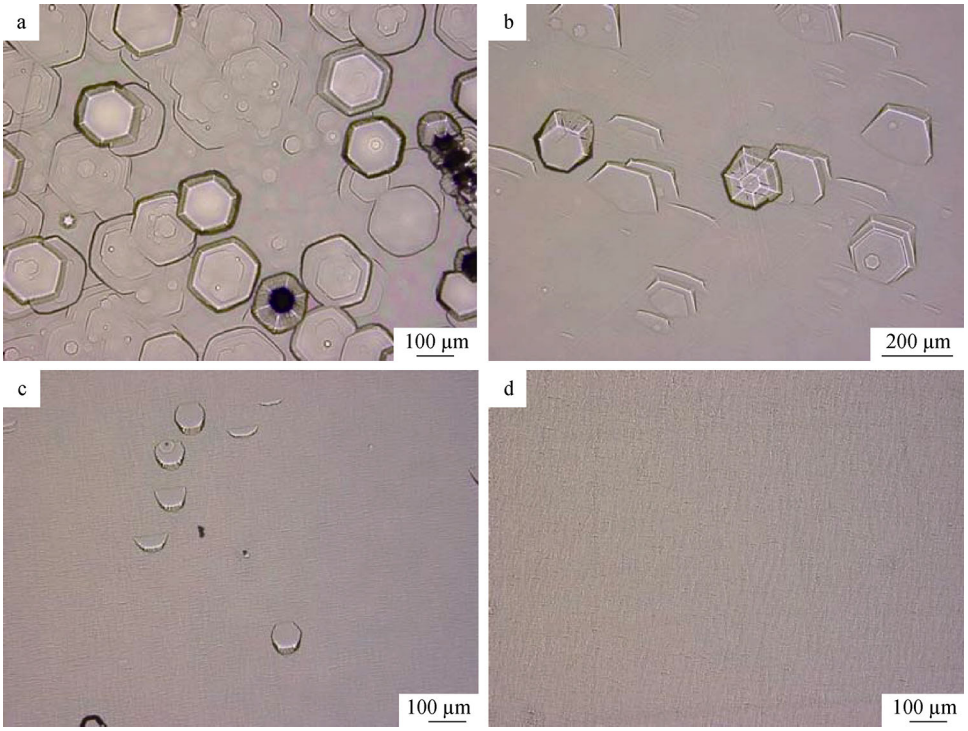


**Fig. 1** OM images of GaN samples grown on sapphire substrate with different mis-orientation angles: **a** on normal sapphire, **b** on 0.5°-off sapphire, **c** on 1.0°-off sapphire, and **d** on 2.0°-off sapphire

realize quantum wells with very sharp interface and benefits the emission efficiency. With the mis-orientation angle increasing, step-bunching is observed in the area free of hexagonal grains. And the surface becomes rougher and rougher due to that the misorientation angle becomes larger. Within each terrace, the surface is still flat but the rms roughness becomes larger with mis-orientation angle increasing. In addition, these steps are along the direction of ⟨1–100⟩ of the GaN.

Figure 3 shows the full widths at half maximum (FWHMs) of XRD ω-rocking curves for symmetric (002) and asymmetric (102) planes for the 2 μm thick N-polar GaN layers grown with different misorientation angles. The FWHM of (002) ω-scans is about 100 arcsec, which is almost not influenced by the mis-orientation except that it is a little bit larger for GaN film with mis-orientation of 2.0°. In addition, the FWHM of (102) ω-scans is about 700 arcsec for GaN layer grown on normal sapphire substrate. It increases to about 1,000 arcsec with the mis-orientation angle increasing to 0.5°, and then gradually decreases with the mis-orientation angle further increasing. It is known that the broadening of (002) ω-scan is mainly due to the threading dislocations (TDs) with screw component (tilt) and the broadening of (102) ω-scan is mainly ascribed to the TDs with edge component (twist). Therefore, it shows in Fig. 3 that GaN film grown at 0.5°-off sapphire shows the highest density of TDs with edge component and the increase of mis-orientation angle of sapphire substrate leads to the reduction of edge-type TDs density.

Figure 4 shows Raman spectra of GaN layers grown on sapphire substrate with different mis-orientation angles, which is measured in back-scattering geometry. As shown in Fig. 4, two peaks are observed for each sample. The dominant peaks are located at around 565 $cm^{-1}$ which is ascribed to $E_2$ (high) mode of GaN. Other peaks located at around 751 $cm^{-1}$ are assigned to $E_g$ mode of sapphire substrate. It is known that the residual strain of epitaxial layers can be estimated from the scattering peak of $E_2$ (high) mode, while the FWHMs of $E_2$ (high) mode scattering peaks can be used as a merit of crystalline quality. Figure 5 shows the Raman peak of $E_2$ (high) mode and its FWHM for the GaN layers grown on sapphire with different mis-orientation angles. It is shown that the Raman frequency of the $E_2$ (high) mode changes from 565.4 to 564.5, 566.0, and 566.7 $cm^{-1}$ for GaN layers grown on normal sapphire substrate and 0.5°, 1.0°, and 2.0° off sapphire substrate. In comparison with the Raman frequency of the $E_2$ (high) mode for strain-free GaN, it is shown that almost all the GaN is undertaken compressive strain, while the sample grown on 0.5°-off sapphire substrate is almost strain free [16]. Then, with the mis-orientation angle further increasing, the compressive strain

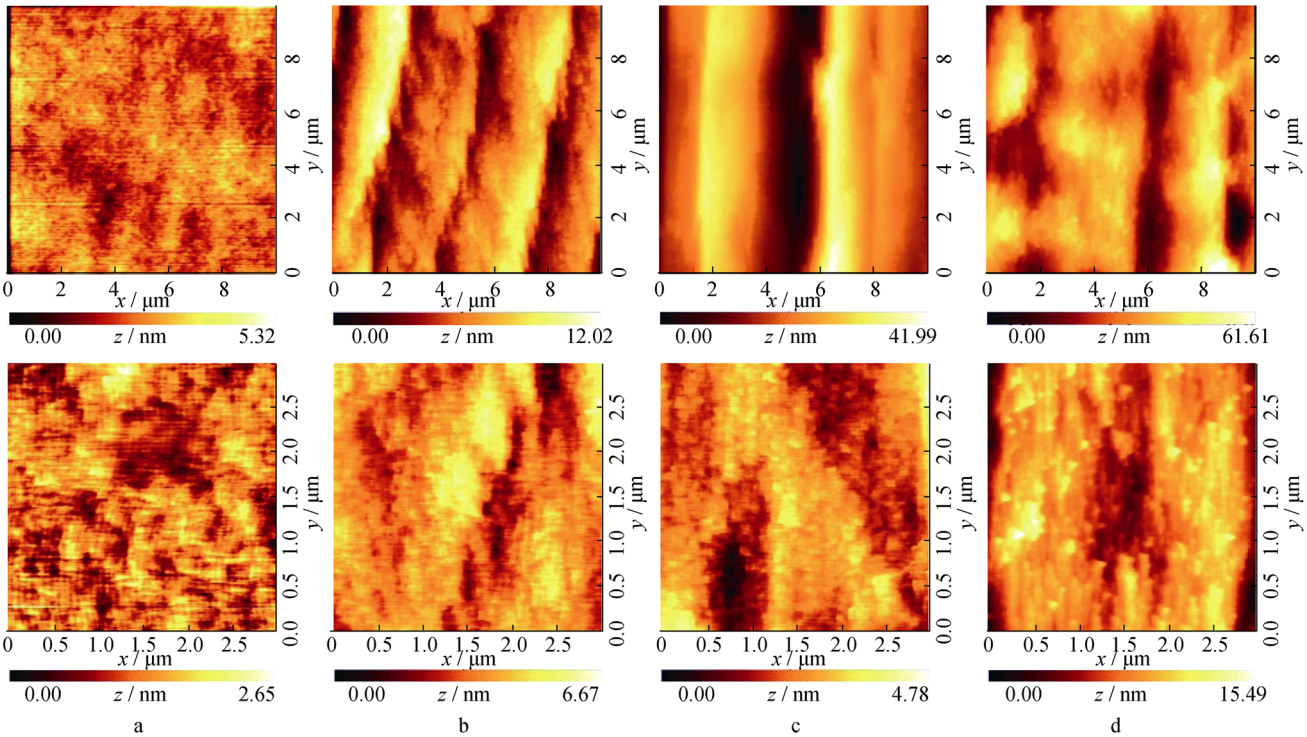


**Fig. 2** AFM images of GaN samples grown on sapphire substrate with different mis-orientation angles: **a** on normal sapphire, **b** on 0.5°-off sapphire, **c** on 1.0°-off sapphire, and **d** on 2.0°-off sapphire. Scanned areas being 10 μm × 10 μm and 3 μm × 3 μm

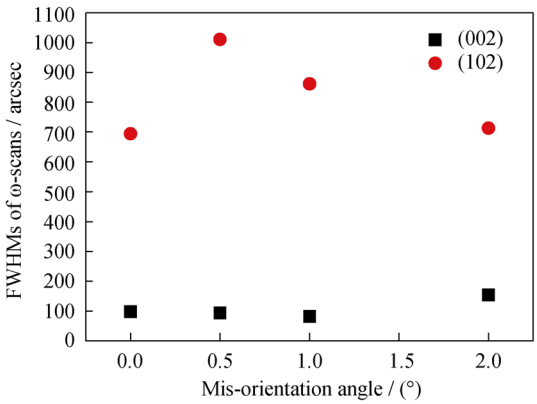


**Fig. 3** FWHMS of ω-rocking curves for (002) symmetric and (102) asymmetric planes of GaN samples grown on sapphire substrate with different mis-orientation angles

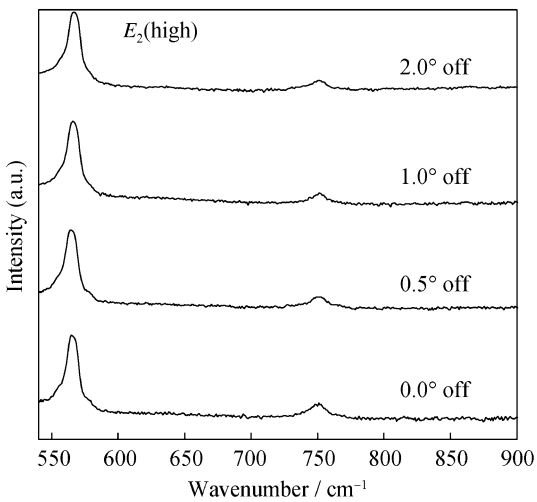


**Fig. 4** Raman scattering spectra of GaN samples grown on sapphire substrate with different mis-orientation angles

becomes larger and larger. In Fig. 5, it is also shown that the FWHMs of the Raman scattering peak of the $E_2$ (high) mode increases from 6.16 to 6.41 cm$^{-1}$, and then decreases to 5.98 cm$^{-1}$ with the mis-orientation angles increasing. This indicates that the crystalline quality of GaN layer becomes worse first and then is gradually enhanced with the mis-orientation angle increasing, which coincides with the tendency of the edge-type TDs as a function of the mis-orientation angle well as shown in Fig. 3. In Fig. 5, it can be seen that the tendency of residual strain and crystalline quality is very similar, i.e., the poorer the crystalline quality is, the smaller the residual strain is. This indicates that the threading dislocation can effectively release the strain in GaN where the GaN with the largest TDs density has the smallest residual compressive strain.

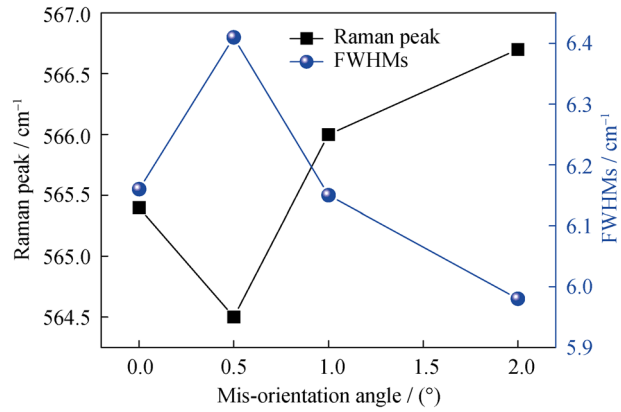


**Fig. 5** Raman scattering peaks of $E_2$ (high) mode and their FWHMs for GaN layers grown on sapphire substrate with different mis-orientation angles

## 4 Conclusion

In summary, N-polarity GaN layers were grown on sapphire substrate by MOVCD. And the flat surface was successful obtained by using the mis-orientation angle sapphire substrate. The hillocks become less and less with the mis-orientation angles increasing and are finally eliminated and resulted in flat surface at the mis-orientation angle of 2.0°. The crystalline quality is firstly reduced for GaN films grown on the mis-orientation angle of 0.5° and then gradually improved with the mis-orientation angle further increasing, as revealed by the XRD and Raman measurement. It is shown in this work that the application of mis-orientation angle substrate is a successful way to obtain the flat GaN with N-polarity and to improve the crystalline quality as well.

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