# Optical and structural investigation of *a*-plane GaN layers on *r*-plane sapphire with nucleation layer optimization*

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Nonpolar *a*-plane GaN epilayers are grown on several *r*-plane sapphire substrates by metal organic chemical vapour deposition using different nucleation layers: (A) a GaN nucleation layer deposited at low temperature (LT); (B) an AlN nucleation layer deposited at high temperature; or (C) an LT thin AlN nucleation layer with an AlN layer and an AlN/AlGaN superlattice both subsequently deposited at high temperature. The samples have been characterized by X-ray diffraction (XRD), atomic force microscopy and photoluminescence. The GaN layers grown using nucleation layers B and C show narrower XRD rocking curves than that using nucleation layer A, indicating a reduction in crystal defect density. Furthermore, the GaN layer grown using nucleation layer C exhibits a surface morphology with triangular defect pits eliminated completely. The improved optical property, corresponding to the enhanced crystal quality, is also confirmed by temperature-dependent and excitation power-dependent photoluminescence measurements.

**Keywords:** *a*-plane GaN, metal organic chemical vapour deposition, AlN/AlGaN superlattice, photoluminescence

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## 1. Introduction

Currently, III-nitride optoelectronics on *c*-sapphire is suffering a strong electric field due to spontaneous and piezoelectric polarization. This results in a reduction in quantum efficiency due to carrier separation and reduced radiative recombination rate. Therefore, nonpolar *a*-plane GaN, which can be achieved on *r*-sapphire, catches a lot of attention in III-nitride based opto-electronic devices, as such self-built electric fields can be eliminated.

However, it is extremely difficult to grow nonpolar GaN on *r*-sapphire due to a number of challenges. Currently, the conventional two-step growth is still the major approach to the growth of non-polar GaN on sapphire, namely, a thin nucleation layer of low temperature (LT) GaN[1] (or high temperature (HT) AlN,[2] multiple layer[3,4]) followed by the growth of an HT GaN layer. It is expected that the thin nucleation layer needs to be optimized in order to improve the crystal quality. In this paper, a number of methods with different nucleation layers have been employed to grow non-polar GaN on *r*-sapphire, showing significant difference in crystal quality and optical property.

## 2. Experiment

The samples were grown on *r*-plane sapphire substrates by vertical low pressure metal organic chemical vapour deposition (MOCVD) under a pressure of 40 Torr ($5.33 \times 10^4$ Pa). The single GaN layer was grown using different nucleation layers (NLs): (A) an LT GaN of about 50-nm thick, denoted as sample A; (B) an HT AlN of about 60-nm thick, denoted as sample B; (C) a 7-nm LT AlN layer with a 60-nm HT AlN layer and AlN/AlGaN superlattice (SL) sequentially deposited, denoted as sample C. In each case, the growth of 1.5-μm GaN epitaxy layer remained under similar conditions. The growth details were similar to those in Refs. [5] and [6].

The high resolution X-ray diffraction (XRD) was

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performed using Bruker D8-discover system equipped with Ge(220) monochromator, delivering a pure Cu K$\alpha$ line of wavelength $\lambda = 0.15406$ nm. The atomic force microscopy (AFM) was performed using an Agilent 5500 scanning probe system. The photoluminescence (PL) measurements were carried out using the 325-nm line of a He–Cd laser and a frequency doubled $Ar^+$ ion laser operating at 244 nm. The samples were held in a helium closed-circuit cryostat in a temperature range from 12 K to 300 K. The luminescence was dispersed using a 0.5-m monochromator and detected using a charge coupled detector (CCD).

## 3. Results and discussion

Generally $a$-plane GaN thin films are characteristic of significant in-plane anisotropy,[5,7–9] and the x-ray rocking curves (XRCs) of $(11\bar{2}0)$ plane measured along different directions show different peak widths. Table 1 lists are the results of full width at half maximum (FWHM) of $(11\bar{2}0)$ XRCs, showing that the GaN layer grown using the thin AlN NL gives a better crystal quality than that using the GaN NL. Furthermore, an introduction of LT AlN and supperlattice can further significantly improve crystal quality of the GaN layer. The $(11\bar{2}0)$ XRC curve along $c$-axis has an FWHM of 674 arcsec and the GaN layer is best among the investigated samples.

Figure 1 shows the surface morphologies of the samples, examined by AFM. Sample A exhibits a large density of small pits generally with a triangular shape. Further experiments indicate that such pits cannot be eliminated only by increasing the film thickness. In the case of sample B (i.e., AlN NL), a fully island coalesced film is not achieved, which might be due to the cracking of HT AlN NL layer under large strain, thereby causing uneven distributions of GaN nuclear sites and grains. Though the growth condition of HT AlN NL needs further optimization, the reduction in FWHM of XRC shown in sample B indicates that the crystal grains should be of higher quality than those in sample A as a result of using the AlN NL. When LT AlN and AlN/AlGaN SLs are introduced below and above the HT AlN layer respectively (sample C), the surface morphology is observed to be free of triangular defect pits. This indicates that the LT AlN plus HT AlN and suplerlattice NL structure is efficient in relieving the lattice and thermal mismatch caused strain and reducing the density of crystal defects, which leads to further improvement on crystal quality. Meanwhile, the surface presents long arrays of almost parallel ridges and valleys with orientations extending toward the $c$-axis. The wavy surface morphologies are widely observed in $a$-plane GaN films,[10] which implies that our next step work should focus on further improving the crystalline quality and achieving mirror-like surface through optimizing the growth condition.

The optical properties of these $a$-plane GaN films were investigated by PL measurements on sample A and sample C. The low temperature PL results measured at 20 K are shown in Fig. 2, where four emission peaks can be found at about 3.47 eV, 3.42 eV, 3.286 eV and 3.195 eV respectively. The physical origin of these peaks was investigated by excitation power dependent PL measurements at 20 K. Only the results of sample A are plotted in Fig. 3, since the emission behaviours of both samples are similar.

**Table 1.** High resolution X-ray diffraction results of $a$-plane GaN samples.

| | Sample A | Sample B | Sample C |
|---|---|---|---|
| NL scheme | LT GaN | HT AlN | LT AlN + HT AlN + SLs |
| $(11\bar{2}0)$ XRC FWHM along $c$-axis/arcsec | 1390 | 1115 | 674 |
| $(11\bar{2}0)$ XRC FWHM along $m$-axis/arcsec | 2407 | 1319 | 1147 |

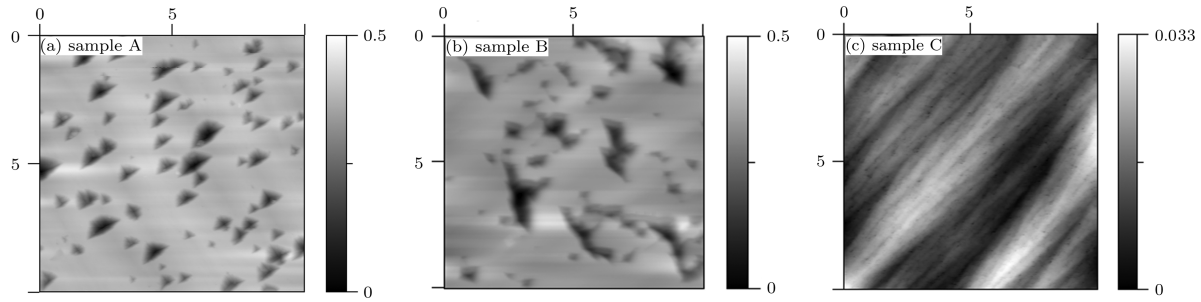



**Fig. 1.** Images of $a$-plane GaN samples observed by atomic force microscopy. The unit in the figures is μm. (a) sample A, (b) sample B, (c) sample C.

The peak at 3.47 eV shows no energy shift when the excitation power changes from 0.125 mW to 20 mW and so is the 3.42 eV peak. In addition, the 3.286 eV peak undergoes a clear blue shift with excitation increasing (10.2 meV for sample A and 9.0 meV for sample C).

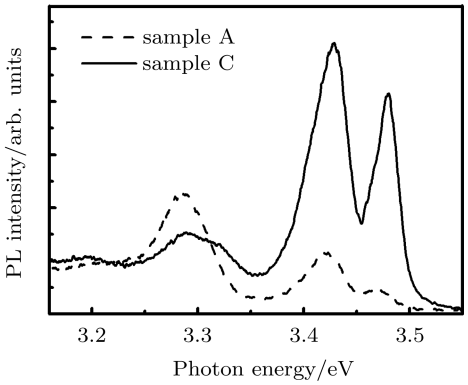



**Fig. 2.** Photoluminescence measured at 20 K.

Therefore, the near-band-edge (NBE) 3.47 eV peak can be ascribed to a donor-bound exciton emission ($D^0X$) and the 3.286 eV peak to a donor–acceptor pair (DAP) recombination emission. Both are similar to those given in other reports.[11−16] The 3.195 eV peak has the same energy shift as the 3.286 eV emission peak with excitation intensity increasing (not shown in Fig. 3) and the energy splitting between these two peaks is kept at about 90 meV. Therefore, it is likely that the 3.195 eV peak might be due to the LO-phonon replica of the 3.286 eV peak. The 3.42 eV peak has been widely observed for *a*-plane films and attributed to the recombination of either excitons/carriers localized to basal stacking-fault (SF)[11−13,17,18] or DAP.[3,19] We are inclined to accept the former as there seems to be no clear energy shift when the excitation intensity increases over three orders of magnitude.

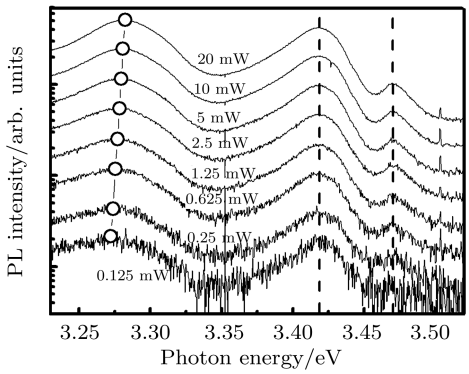



**Fig. 3.** Excitation power dependent PL spectra of sample A with the excitation intensity increasing from 0.125 mW to 20 mW at 20 K. The dashed lines demonstrate no peak shift occurring to the peaks at 3.47 and 3.42 eV. The symbolized line shows a blue shift happening to the peak at 3.286 eV.

Having in mind the assignment of these emission peaks, the optical property of the samples, presented in Fig. 2 and the temperature behaviour of PL in Fig. 4, can be compared and it can be found that they are closely related to the crystalline property.

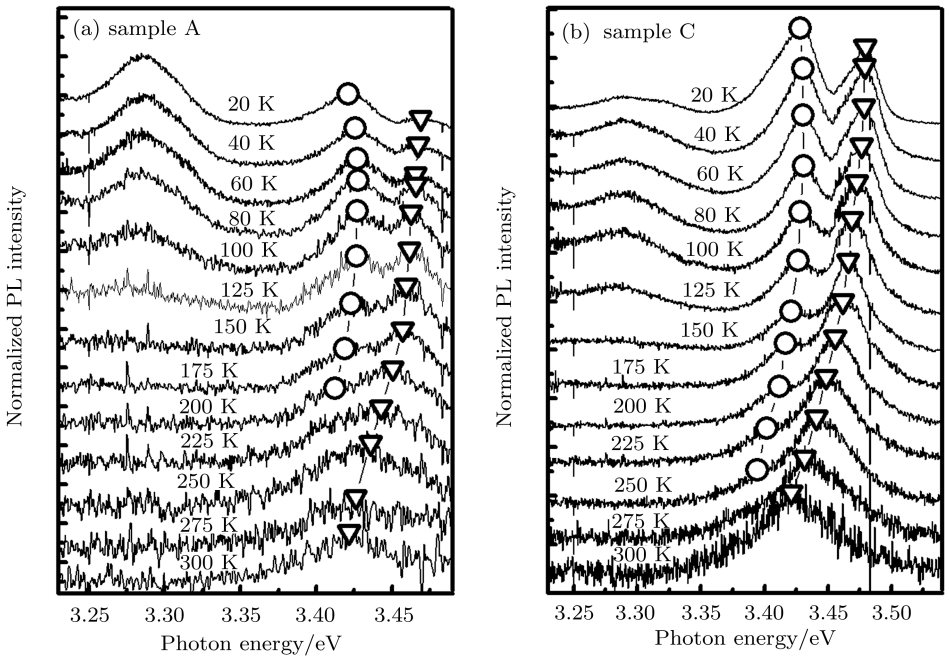



**Fig. 4.** Temperature dependences of PL spectra from 20 K to room temperature. The energy positions shifts happen up to 3.47 and 3.42 eV peaks are indicated by symbolized lines. (a) sample A, (b) sample C.

From Fig. 2 it can be seen that in sample C the 3.47 eV NBE emission is stronger and narrower and the 3.286 eV DAP emission is weaker. These features clarify the improved optical property of sample C compared with that of sample A. The 3.42 eV peak, based on its origin from SFs, has been reported to have a higher emission intensity for samples with fewer but larger SFs and a lower emission intensity for samples with more but smaller SFs.[18] So the higher peak for sample C hints at better crystalline integrity, i.e. improved crystal quality. In addition, the 3.42 eV peaks in Fig. 4 for both samples each show a blueshift when temperature increases from 20 K to 80 K, and then a redshift above 80 K, indicating carrier localization in SFs. The blueshift energy is 6.23 meV for sample A and 2.55 meV for sample C, which means that the localization is stronger for sample A,[11] a fact that is also consistent with the case where sample A contains more SFs. The thermal quenching effects occur to all the peaks in Fig. 4 at elevated temperature. The 3.286 eV peaks disappear above 150 K for both samples, which is in accordance with those given in other reports[14,15] for $a$-plane GaN films. However, in sample C the NBE emission maintains far better intensity and narrower peak shape up to the room temperature and the SF emission is better resolved above 150 K, which also exhibit the enhanced optical property for sample C. Therefore, it can be concluded that the NL composed of LT AlN plus HT AlN and suplerlattice can lead to improved crystal quality and thus better optical property of the non-polar GaN.

## 4. Conclusion

Nonpolar $a$-plane GaN epilayers on $r$-plane sapphire substrates grown by MOCVD with different nucleation layers are investigated by HRXRD, AFM and PL. The measured XRD rocking curves and the surface morphology indicate that the HT AlN NL can give better crystal quality than the LT GaN NL and the multilayer NL of LT AlN plus HT AlN and suplerlattice can significantly improve GaN crystal quality. The PL results reveal the close relation between the optical property and the crystalline property of $a$-plane GaN films and support strongly the effectiveness of the LT AlN plus HT AlN and suplerlattice NL structure.

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