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Optimization of (1 1 $\bar{2}$ 0) a-plane GaN growth by MOCVD on (1 $\bar{1}$ 0 2) r-plane sapphire

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Abstract

A-plane GaN (1 1 $\bar{2}$ 0) epilayers have been grown on r-plane (1 $\bar{1}$ 0 2) sapphire by MOCVD, and investigated by scanning electron microscopy (SEM), X-ray diffraction (XRD), and atomic force microscopy (AFM). This particular orientation is non-polar, as opposed to the c-direction, and avoids polarization charge and the associated screening charge, and the consequent band bending. Our results showed that low pressure, low ammonia flow rate (namely low V/III ratio), high-temperature conditions lead to fully coalesced and relatively high crystalline-quality a-plane GaN films. Both low-temperature GaN buffer and high-temperature AlN buffer were used for a-plane GaN growth on r-plane sapphire, and produced similar crystalline quality and surface morphology. Surface morphological evolution during early stages of a-plane GaN growth revealed behavior different from that of c-plane GaN growth. The possible reasons for striped features and surface undulations of a-plane GaN are discussed.

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Keywords: A3. Metalorganic chemical vapor deposition; B1. Nitrides; B1. Semiconducting III–V materials

1. Introduction

The great majority of research on GaN involves the basal plane, c-plane, wurtzite epilayers. In the c-orientation, the internal spontaneous and strain-induced piezoelectronic polarization effects produce strong electric fields at the heterointerfaces due to the former and everywhere due to the latter. Although the electric field can be advantageous for two-dimensional electron gas formation in FETs without external doping, but it causes spatial separation of electrons and holes in quantum wells of light-emitting diode (LED). Such a separation increases the radiative lifetime [1] at the expense of the quantum efficiency [2]. It also causes a red shift in LEDs and makes the emission wavelength dependent on injection unless very thin quantum wells are employed. In short, additional constraints are placed on design rules in an effort to deal with polarization-induced field. One approach to overcome this problem is to grow m-plane or a-plane hexagonal GaN that is not polar. Since the reported demonstration of growth of a-plane GaN by MBE [3] and MOCVD [4], much attention has been focused on a-plane GaN growth. Up to now, epitaxial lateral overgrowth (ELOG, also goes by the nomenclature epitaxial lateral overgrowth—ELO) of a-plane GaN [5,6] and related LED [7] have been reported. However, reports on detailed and systematic investigations of growth of a-plane GaN are lacking. In this paper, we explored the parameter space for the optimization of a-plane GaN growth on r-plane sapphire by MOCVD including various buffer layer schemes.

2. Experimental procedure

(1 1 $\bar{2}$ 0) a-plane GaN films were grown on (1 $\bar{1}$ 0 2) r-plane sapphire (with $\pm 2^{\circ}$ surface tolerance). Prior to

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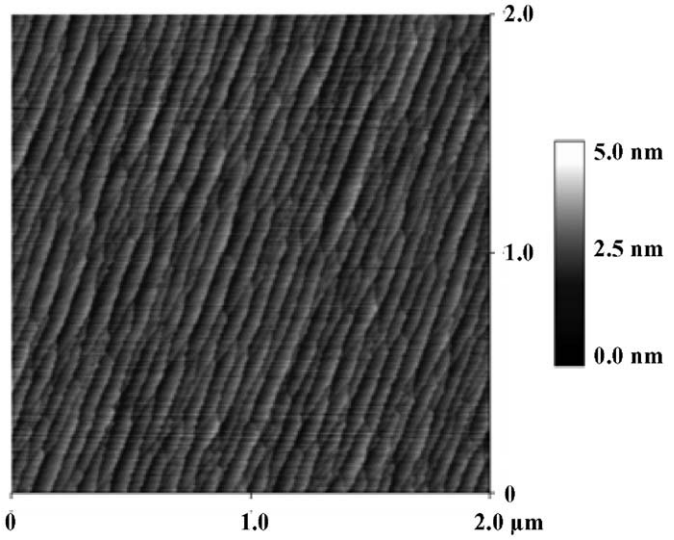

Fig. 1. AFM image for annealed r-plane (1 $\bar{1}$ 0 2) sapphire substrate, with RMS value of 0.36 nm.

growth, the sapphire substrates were annealed at 1400 °C for 1 h in air ambient with annealing furnace to generate atomic steps on substrate surface, as shown in Fig. 1. After chemical cleaning, sapphire substrates, one at a time, were loaded into the low-pressure custom-designed organo-metallic vapor phase epitaxy (OMVPE) system. Following in situ annealing of sapphire, a low-temperature (LT) GaN buffer layer was deposited at 550 °C with a thickness of 45 nm. In the case of using high-temperature (HT) AlN as buffer layer, the buffer layer growth temperature and thickness were 1080 °C and around 100 nm, respectively. After buffer layer growth, the temperature was raised to a value in between 1040 and 1070 °C, and a-plane GaN epilayer was grown subsequently. Trimethylgallium (TMGa), trimethylaluminum (TMAl) and ammonia were used as the Ga, Al and N sources, respectively. The as-grown samples were characterized by scanning electron microscopy (SEM), atomic force microscopy (AFM), and high-resolution X-ray diffraction.

3. Results and discussion

The growth parameter space for optimization included chamber pressure, ammonia flow rate, and epilayer growth temperature. Fig. 2 shows the effect of growth pressure on a-GaN surface morphology. During optimization, 0.9 μm-thick GaN films were grown under two different growth pressures: 30 and 200 Torr. As shown in Fig. 2(b), the lateral growth rate under 200 Torr was rather small with respect to the vertical growth rate, and consequently we could hardly observe the presence of flat fully coalesced area on the sample surface. By comparison, a-plane GaN grown under 30 Torr (Fig. 2(a)) showed a better tendency of lateral growth, albeit with large pits decorating the surface. The pits were very stable under unoptimized growth conditions, and could not be eliminated by simply

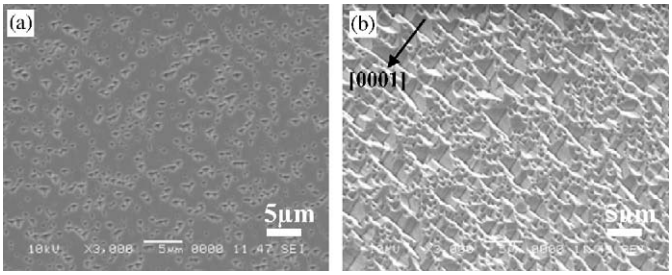

Fig. 2. SEM images of 0.9 μm a-plane GaN (1 1 $\bar{2}$ 0) grown on r-plane (1 $\bar{1}$ 0 2) sapphire at 980 °C, with 7000 ccm NH_3, under (a) 30 Torr, (b) 200 Torr, respectively.

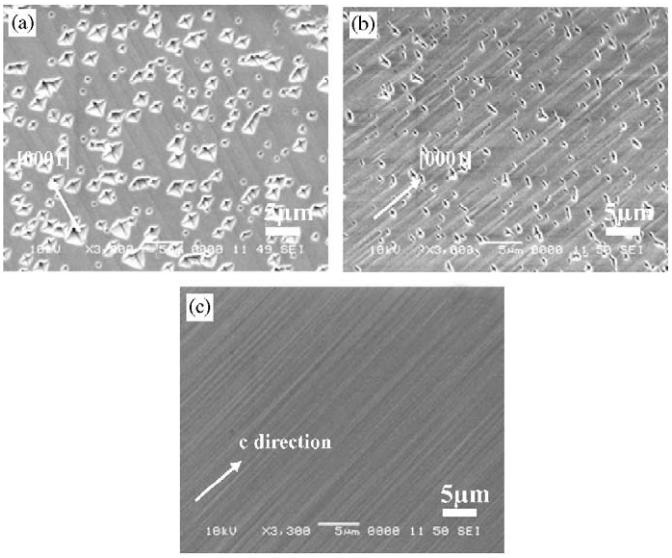

Fig. 3. SEM images of a-plane GaN (1 1 $\bar{2}$ 0) grown under 30 Torr, at 1070 °C, with NH_3 flow rates of (a) 7060 ccm, (b) 4500 ccm, (c) 1000 ccm, respectively.

increasing the film thickness. Wu et al. [8] reported that these triangular pits were composed of one {0 0 0 $\bar{1}$} and two {1 0 $\bar{1}$ 1} facets.

Ammonia flow rate is also an important parameter for growth of a-plane GaN. Fig. 3 shows the effect of NH_3 flow rate on the surface morphology of a-plane GaN. The three samples shown here have the same thickness of around 1.5 μm, with different-sized pits on the surface. With decreasing flow rate of NH_3, the size of the triangular pits was reduced from several microns to zero. Therefore, a relatively low V/III ratio was found to be beneficial in reducing the size of pits on the sample surface and attaining coalesced GaN films. Additionally, we can determine [0 0 0 1] or [0 0 0 $\bar{1}$] direction according to stripes and the shape of triangular pit on surface [8], as shown in Fig. 3.

We have also investigated the influence of the growth temperature on a-plane GaN, as shown in Fig. 4. The pit size was reduced with increasing growth temperature from 1050 to 1090 °C. However, from these SEM images, we can also see that the surface undulation became more significant with increasing growth temperature, which was

also confirmed by surface roughness measurements using AFM, which are not shown here. The full width at half maximum (FWHM) value of the X-ray rocking curve of GaN ($1\,1\,\bar{2}\,0$) showed in-plane anisotropy, and was a function of the in-plane azimuth angle, consistent with the report by Wang et al. [9]. The FWHM values of X-ray rocking curve were 0.46°–0.67°, 0.29°–0.43°, and 0.28°–0.43° for samples grown at 1050, 1070, and 1090 °C, respectively. Higher growth temperature could yield a-plane GaN with improved crystalline quality. Therefore, the choice of optimum temperature should be a compromise of surface morphology and crystalline quality.

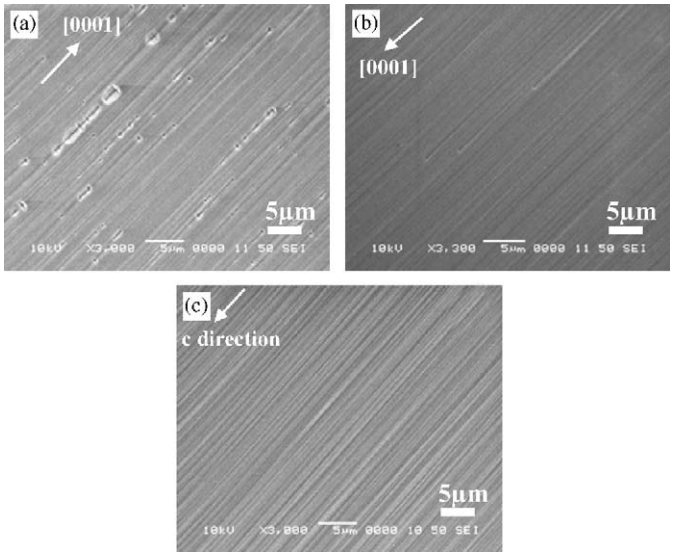

Fig. 4. SEM images of a-plane GaN ($1\,1\,\bar{2}\,0$) grown under 30 Torr with NH_3 flow rate of 1000 sccm, with growth temperature being (a) 1050 °C, (b) 1070 °C, (c) 1090 °C, respectively.

Fig. 5 shows the surface morphological evolution during early stage of a-plane GaN growth. These SEM images show that the 45 nm LT-GaN buffer was featureless, as shown in Fig. 5(a). Continuing on with further growth, the 20 nm-thick GaN in Fig. 5(b) was dominated by nucleation islands. When the thickness increased to around 50 nm (Fig. 5(c)), the islands began to coalesce and form elongated features running along one direction. We determined this to be the c-direction of GaN. With the growth continuing, the elongated islands started to merge with each other, as shown in Fig. 5(d). For the 300 nm-thick GaN (Fig. 5(e)), the sample was almost fully coalesced, with some small pits on the surface. When the thickness reached 1.5 μm (Fig. 5(f)), the film fully coalesced. From this image, a-plane GaN showed stripe features along the c-direction, and surface undulation along the m-direction ([$1\,\bar{1}\,0\,0$]) as well, which is perpendicular to the c-direction ([$0\,0\,0\,1$]). From the elongated island growth, we can also conclude that the growth rate along the c-direction is much higher than that along the m-direction. It is worth mentioning here that by performing lateral overgrowth using our optimized growth conditions, the ratio of the lateral growth rate along the c-direction to the vertical growth rate was around 3–4, which is relatively higher than those reported in the literature [5,6]. Additionally, from the growth evolution, it is suggested that at the very beginning of a-plane GaN epitaxy, the growth follows a Volmer–Weber (VW) mode, which gives way to a Frank–van der Merwe (FM) mode after most of the islands have merged to each other.

In order to further optimize a-plane GaN films, we have also investigated the effect of HT-AlN buffer layers in addition to LT-GaN buffers, which was used for the above-mentioned a-plane GaN samples. The AlN buffer layer was 100 nm thick, and was grown at 1080 °C using

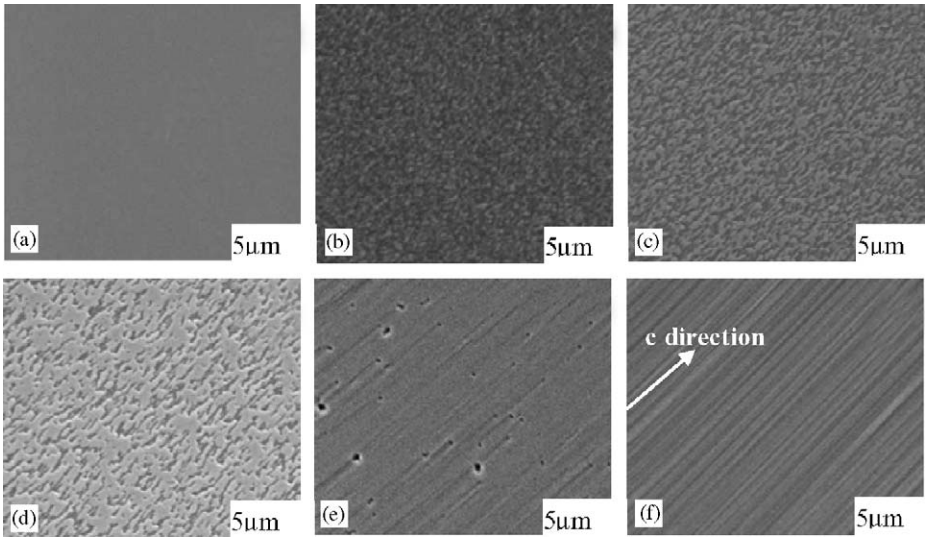

Fig. 5. Surface morphological evolution during early stage of a-plane GaN growth: (a) LT-GaN buffer, (b) 20 nm GaN, (c) 50 nm GaN, (d) 100 nm GaN, (e) 300 nm GaN, (f) 1.5 μm fully coalesced GaN. Note that c-direction is the stripe direction shown in (f), and all samples here have their c directions parallel to each other.

TMAl as the Al source. The flow rates for TMAl, and NH_3 were 10 μmol/min, and 7 ccm, respectively. A-plane GaN epilayers were grown at 1070 °C, 30 Torr, with 1000 ccm NH_3. X-ray diffraction (XRD) measurements with $2\theta-\omega$ scan (see Fig. 6) showed $(2\bar{2}04)$ r-plane of sapphire, $(11\bar{2}0)$ of AlN and $(11\bar{2}0)$ of GaN parallel to each other. The FWHM value of the X-ray rocking curve for $(11\bar{2}0)$ GaN was in the range of 0.29°–0.52°. AFM and SEM measurements were used to characterize the surface morphology of these a-plane GaN films, the results of which are shown in Fig. 7. AFM root mean square (RMS) roughness was about 15 nm, similar to that of a-plane GaN grown at the same temperature using a LT-GaN buffer. An SEM image revealed fully coalesced surface and stripe features along the c-direction, which was also observed in the case of LT-GaN buffer layers.

From the above experimental results, stripes features along the c-direction and surface undulation along the m-direction were found on all a-plane GaN samples, no matter what kind of buffer layer was used for epitaxy. First, we can rule out the possibility that the surface undulation comes from the atomic steps of annealed r-plane sapphire formed during annealing as the same surface features were obtained on unannealed sapphire substrates, which does not have those atomic steps on its surface. One possible explanation is that the surface undulation may be caused by the disparity of migration lengths of adatoms along the two perpendicular directions, c- and m-directions, during growth, as shown in the case of high-index GaAs surfaces such as GaAs (3 3 1) [10]. This disparity of migration length could be enhanced with increasing temperature, therefore resulting in more significant surface undulation for a-plane GaN films at elevated growth temperature.

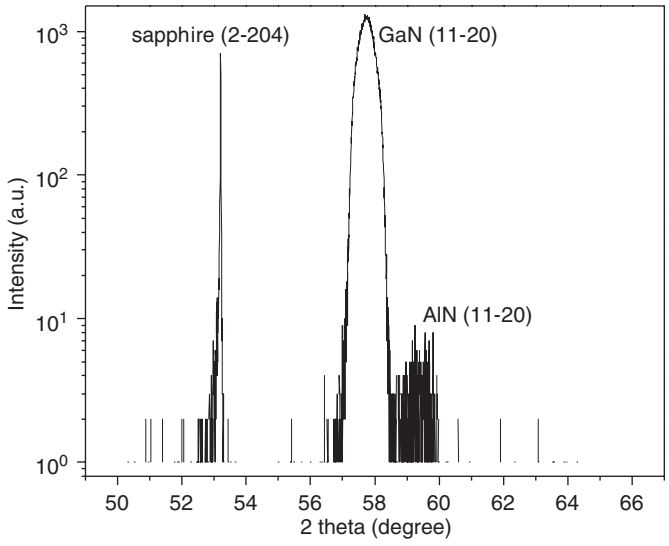

Fig. 6. XRD spectra using $2\theta-\omega$ scan for a-plane GaN grown with LT-AlN buffer layer on r-plane $(1\bar{1}02)$ sapphire.

4. Conclusions

A-plane GaN growth conditions by MOCVD, including the chamber pressure, ammonia flow rate and epilayer growth temperature, have been optimized using LT-GaN buffer layer. Low pressure, low ammonia flow rate (namely low V/III ratio), high growth temperatures have been found to be beneficial for the growth of fully coalesced and relatively higher crystalline-quality a-plane GaN films. We also explored a-plane GaN epilayer using HT-AlN buffer layer, with similar crystalline quality and surface morphology to a-plane GaN using LT-GaN buffer. The possible reasons for striped features and surface undulation observed on the as grown a-plane GaN were discussed. In general, the quality of a-plane GaN $(11\bar{2}0)$ on r-plane $(1\bar{1}02)$ sapphire obtained in our laboratory as well as others is inferior to that on c-plane sapphire. One of the reasons must certainly have to do with enormous lattice distortion that takes place in transitioning from r-plane sapphire to a-plane GaN. The actual bonding configuration remains to be determined.

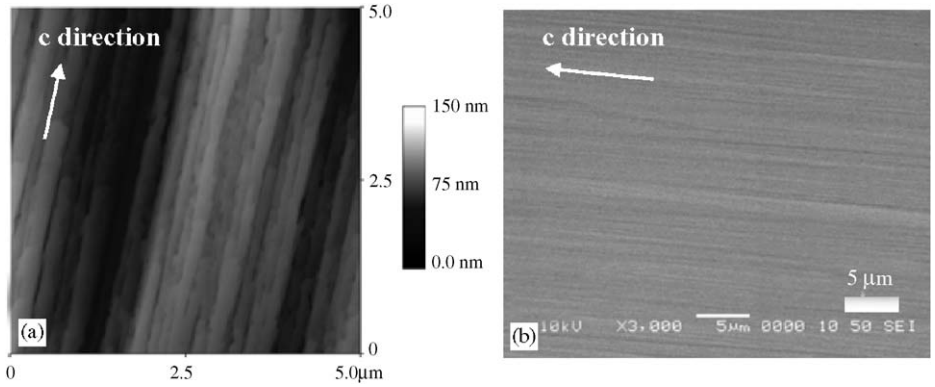

Fig. 7. Surface morphology measurements of a-plane GaN $(11\bar{2}0)$ films grown with HT-AlN buffer, by (a) AFM and (b) SEM measurements.

References

[1] R. Langer, J. Simon, V. Ortiz, N.T. Pelekanos, A. Barski, R. Andre, M. Godlewski, Appl. Phys. Lett 74 (1999) 3827.
[2] T. Deguchi, K. Sekiguchi, A. Nakamura, T. Sota, R. Matsuo, S. Chichibu, S. Nakamura, Jpn. J. Appl. Phys. Part 2—Lett. 38 (8B) (1999) L914.
[3] H.M. Ng, Appl. Phys. Lett. 80 (2002) 4369.
[4] M.D. Craven, S.H. Lim, F. Wu, J.S. Speck, S.P. DenBaars, Appl. Phys. Lett. 81 (2002) 469.
[5] M.D. Craven, S.H. Lim, F. Wu, J.S. Speck, S.P. DenBaars, Appl. Phys. Lett. 81 (2002) 1201.
[6] C.Q. Chen, J.W. Yang, H.M. Wang, J.P. Zhang, V. Adivarahan, M. Gaevski, E. Kuokstis, Z. Gong, M. Su, M.A. Khan, Jpn. J. Appl. Phys. Part 2—Lett. 42 (6B) (2003) L640.
[7] A. Chitnis, C. Chen, V. Adivarahan, M. Shatalov, E. Kuokstis, V. Mandavilli, J. Yang, M.A. Khan, Appl. Phys. Lett. 84 (2004) 3663.
[8] F. Wu, M.D. Craven, S. Lim, J.S. Speck, J. Appl. Phys. 94 (2003) 942.
[9] H.M. Wang, C.Q. Chen, Z. Gong, J.P. Zhang, M. Gaevski, M. Su, J.W. Yang, M.A. Khan, Appl. Phys. Lett. 84 (2004) 499.
[10] H.-P. Schönherr, J. Fricke, Z. Niu, K.-J. Friedland, R. Nötzel, K.H. Ploog, Appl. Phys. Lett. 72 (1998) 566.