High quality AlGaN grown on a high temperature AlN template by MOCVD*

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Abstract: A high temperature AlN template was grown on sapphire substrate by metalorganic chemical vapor deposition. AFM results showed that the root mean square of the surface roughness was just 0.11 nm. Optical transmission spectrum and high resolution X-ray diffraction (XRD) characterization both proved the high quality of the AlN template. The XRD (002) rocking curve full width at half maximum (FWHM) was about 53.7 arcsec and (102) FWHM was about 625 arcsec. The densities of screw threading dislocations (TDs) and edge TDs were estimated to be $\sim 6 \times 10^6$ cm^{-2} and $\sim 4.7 \times 10^9$ cm^{-2}. AlGaN of Al composition 80.2% was further grown on the AlN template. The RMS of the surface roughness was about 0.51 nm. XRD reciprocal space mapping was carried out to accurately determine the Al composition and relaxation status in the AlGaN epilayer. The XRD (002) rocking curve FWHM of the AlGaN epilayer was about 140 arcsec and (102) FWHM was about 537 arcsec. The density of screw TDs was estimated to be $\sim 4 \times 10^7$ cm^{-2} and that of edge TDs was $\sim 3.3 \times 10^9$ cm^{-2}. These values all prove the high quality of the AlN template and AlGaN epilayer.

Key words: AlN template; high temperature; AlGaN; MOCVD; XRD
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1. Introduction

AlGaN-based ultraviolet light-emitting diodes (UV-LEDs) are expected to be used in many applications, such as the curing of various materials (photopolymerization), screen printing, dermatology-related sensing and cures, DNA analyzers, air purification, water disinfection and sterilizers[1−4]. It is widely considered that UV-LEDs have great commercial potential in excess of several billion dollars[5]. When the Al composition changes from 0 to 1, the bandgap of the AlGaN material varies from 3.43 eV (GaN) to 6.04 eV (AlN), while ultraviolet light of wavelengths from 365 to 210 nm can be emitted[6−9]. One key issue of making UV-LEDs is the growth of high quality AlGaN material. Compared with GaN, the growth of high quality AlGaN material has proved to be much more difficult, because of its intrinsic properties. In the commonly used two-step MOCVD growth process, an AlGaN layer is often directly grown on a low temperature (LT) AlN buffer layer. The crystalline quality of AlGaN deteriorates as the Al composition increases[10]: the higher the Al composition, the lower the quality of the AlGaN epilayer. There have been many reports of high quality AlGaN grown on a GaN template. A GaN template indeed effectively improves the quality of the AlGaN epilayer thereon, but it is not suitable for AlGaN with high Al composition. As the lattice parameter of AlGaN is smaller than that of the GaN template, the AlGaN epilayer is under biaxial tensile strain, and crack networks generate when the thickness of the AlGaN layer exceeds the critical value. Methods have been developed to avoid cracks[10−12]. However, in these reports, the Al composition in AlGaN is not high. For AlGaN with high Al composition, this problem is still very tough. Besides, GaN templates are not good choices for deep UV-LEDs, because they would strongly absorb the UV light. Therefore better ways should be developed to grow high quality AlGaN with high Al composition. In this paper, we report our recent research work on MOCVD growth of high quality AlGaN by using a high temperature AlN template.

2. Experimental procedure

The HT-AlN template was grown on 50 mm diameter (0001)-oriented sapphire substrates. Trimethylgallium (TMGa), trimethylaluminum (TMAl) and NH_3 were used as Ga, Al and N precursors. NH_3 flow rates ranged from 1 to 3.5 slpm. H_2 and N_2 were used as the carrier gas. Prior to growth, the substrates were treated in H_2 ambient at 1080 °C for about 6 min. After that, the temperature was decreased to about 600 °C to grow a thin AlN buffer layer. This layer is about 25 nm in thickness. Then the HT-AlN template was grown at a nominal temperature of 1200 °C for 1 h. The growth rate was about 1 μm/h. The AlGaN layer was further grown on the HT-AlN template layer and the growth temperature was about 1100 °C. The AlGaN layer was about 0.8 μm in thickness. The growth pressure in the MOCVD reactor chamber was kept at 50 Torr.

After growth, samples were characterized by UV-VIS optical transmission spectrometry, a Veeco Nanoscope Dimension 3100 atomic force microscopy (AFM) system, and a Bede

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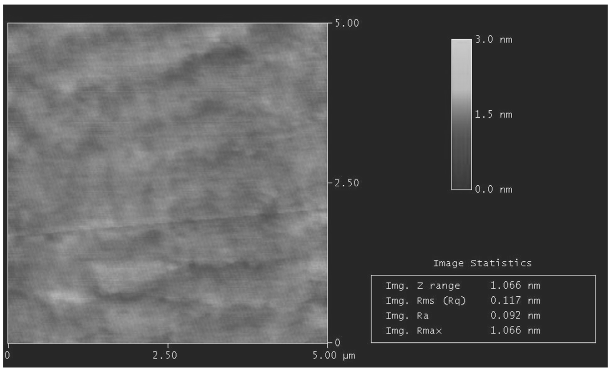

Fig. 1. AFM images of the HT-AlN template.

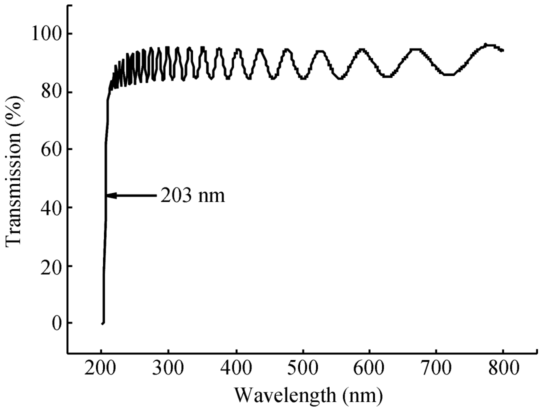

Fig.2. Optical transmission spectrum of HT-AlN template.

D1 high resolution X-ray diffraction (HRXRD) system.

3. Results and discussion

First we studied the surface morphology of the AlN template by AFM. An AFM image is shown in Fig. 1. The root mean square (RMS) of the surface roughness was about 0.11 nm. This value is very small, which means that the surface of the AlN template was very smooth. The smooth surface is considered to benefit from a high growth temperature. Al adatoms have a much larger sticking coefficient than Ga adatoms. Al adatoms are much less mobile on the surface; they tend to cause islands to nucleate, rather than incorporating at the most energetically favorable lattice sites such as a step[4]. At a high temperature of 1200 °C, the surface mobility of Al adatoms can be effectively boosted, and the surface morphology of AlN is thereby improved.

Optical transmission spectra are very useful for material characterization[13, 14]. Optical transmission of the HT-AlN template was measured as a function of wavelength, as can be seen in Fig. 2. The slope of the band-edge is usually indicative of the material quality: the higher the slope, the better the material quality. One can see from Fig. 2 that the absorption band-edge of our sample is very sharp, which indicates the good quality of the AlN template. The band-edge is at about 203 nm. Transmission fluctuation can be seen in the figure, which is caused by optical interference and is related to AlN film thickness. From the position of these interference peaks, we determined the thickness of the HT-AlN template to be about 1 μm.

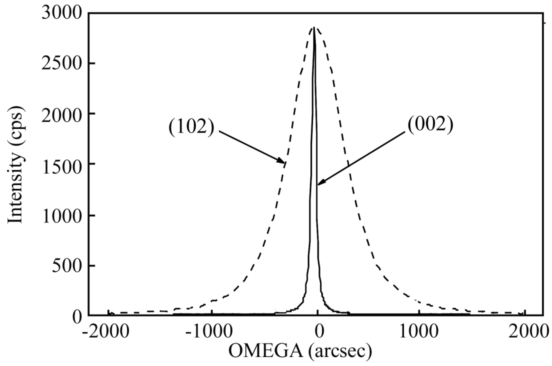

Fig. 3. XRD (002) and (102) rocking curves of HT-AlN template.

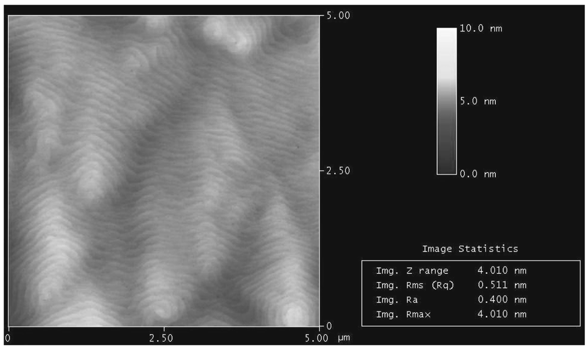

Fig. 4. AFM images of AlGaN grown on HT-AlN template.

The AlN template was further characterized by HRXRD. XRD ω scan rocking curves are shown in Fig. 3. The solid line corresponds to (002) scan, and the FWHM is about 53.7 arcsec. The dashed line corresponds to (102) scan, and the FWHM is about 625 arcsec. From the XRD FWHM values, the densities of screw threading dislocations (TDs) D_S and edge TDs D_E were estimated based on the following equations[15]:

$$D_S = \frac{\beta_S^2}{4.35\,|\boldsymbol{b}_S|^2}, \tag{1}$$

$$D_E = \frac{\beta_E^2}{4.35\,|\boldsymbol{b}_E|^2}, \tag{2}$$

where $|\boldsymbol{b}_S|$ and $|\boldsymbol{b}_E|$ are the Burgers vector sizes of the screw TDs ($|\boldsymbol{b}_S|$ = 0.4982 nm) and edge TDs ($|\boldsymbol{b}_E|$ = 0.3112 nm). β_S and β_E are estimated using the method outlined previously by Lee *et al.*[16]:

$$\beta = \sqrt{(\beta_S \cos\alpha)^2 + (\beta_E \cos\alpha)^2}, \tag{3}$$

where α is the angle between the reciprocal lattice vector (K_{hkl}) and the (001) surface normal. D_S was calculated to be $\sim 6 \times 10^6$ cm^{-2} and D_E was $\sim 4.7 \times 10^9$ cm^{-2}. These values also prove the high quality of the AlN template.

The surface morphology of AlGaN grown on the HT-AlN template was characterized by AFM. Figure 4 shows the AFM image. The RMS of the surface roughness was about 0.51 nm. Though this value is a little larger than that of the AlN template, it is still small enough to prove that the AlGaN epilayer has an atomic-level smooth surface. “Step flow” evidence is

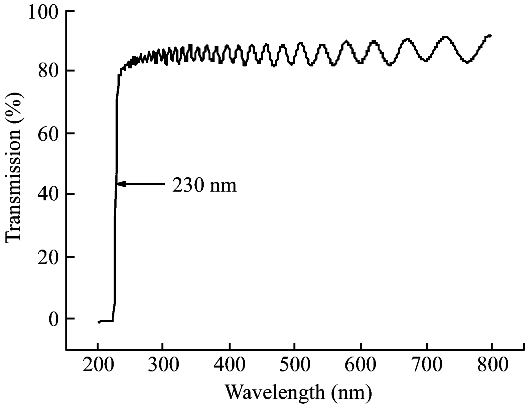

Fig. 5. Optical transmission spectrum of AlGaN grown on HT-AlN template.

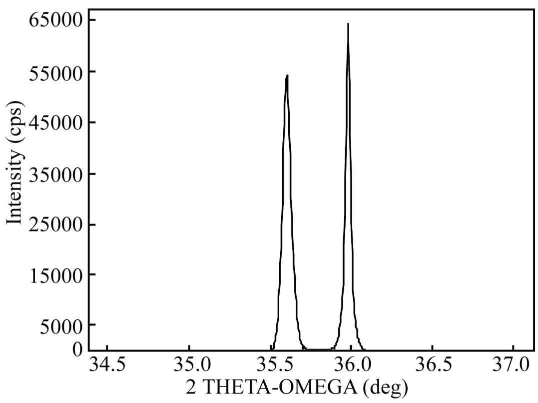

Fig. 6. HRXRD symmetric (002) ω–2θ scan of AlGaN grown on HT-AlN template.

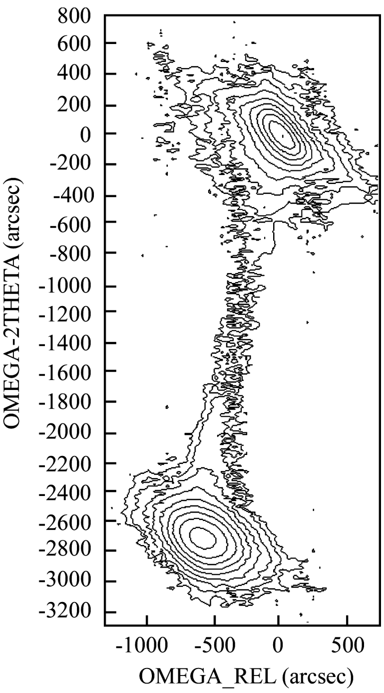

Fig. 7. (105) XRD asymmetric reciprocal mapping of AlGaN grown on AlN template.

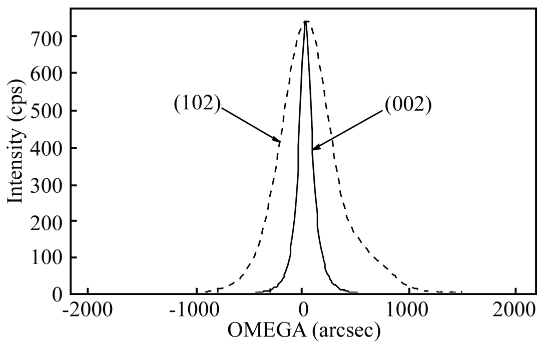

Fig. 8. XRD (002) and (102) rocking curves of AlGaN epilayer.

clearer in this image compared to the AlN template, which means that the growth mode of AlGaN is pretty good[17–19].

The optical transmission spectrum of the AlGaN layer is shown in Fig. 5. As can be seen, the band-edge is at about 230 nm. For $Al_xGa_{1-x}N$ system material, the band-gap energy E_g can be determined with the relationship

$$E_g(x) = 6.04x + 3.43(1-x) - bx(1-x), \tag{4}$$

which has been adopted by many groups working on this material system; b is the so-called bowing parameter[4]. For $b = 1$, with the band-edge wavelength, we determined the value of x to be 81%. The band-edge here is also very abrupt, and therefore the crystalline quality of the AlGaN layer is very good.

Figure 6 shows the XRD symmetric (002) ω–2θ scan result of AlGaN grown on the HT-AlN template. Two strong peaks can be seen in the figure. The left peak is related to the X-ray diffraction of the AlGaN layer, while the right one is related to the HT-AlN template. From the position of the AlGaN peak, we determined the AlGaN layer's Bragg angle to be 17.8°. With Bragg's formula

$$2d\sin\theta = n\lambda, \tag{5}$$

we can obtain the value of the c axial lattice parameter of AlGaN. Using Vegard's law, we can further determine the Al composition to be 79.2%, which is a little different from the value obtained from the optical transmission spectrum before. The reason for this discrepancy may lie in the relaxation of the AlGaN epilayer. The result of 79.2% is based on the assumption that the AlGaN layer is fully relaxed. As a matter of fact, AlGaN layer is merely 0.8 μm in thickness and is therefore not thick enough to fully relax. Besides, the bowing parameter b may not be precisely 1.

A more accurate composition and relaxation status for the AlGaN epilayer can be extracted from asymmetrical XRD reciprocal mapping. In Fig. 7 we show the (105) asymmetrical XRD reciprocal mapping result. The top AlN template reciprocal lattice point is not in vertical alignment with the AlGaN epilayer, indicating that AlGaN was not pseudomorphically grown on the AlN template. We calculated the lattice constants a and c for the AlGaN epilayer, respectively, to be 3.1166 Å and 5.0329 Å. The Al composition in AlGaN is 80.2%, and the relaxation is 30.3%.

XRD ω scan rocking curves of AlGaN are shown in Fig. 8. The solid line corresponds to (002) scan, the FWHM is about 140 arcsec. The dashed line corresponds to (102) scan, the FWHM is about 537.4 arcsec. According to Eqs. (1)–(3), the density of screw TDs D_S was estimated to be $\sim 4 \times 10^7$ cm^{-2} and that of edge TDs D_E was $\sim 3.3 \times 10^9$ cm^{-2}. These values further prove the high quality of the AlGaN epilayer grown on the AlN template.

4. Conclusion

In the paper, we report our research work on AlGaN grown on an HT-AlN template by MOCVD. The AlN template was grown on sapphire substrate at 1200 °C. AFM results showed that the RMS of the surface roughness was just 0.11 nm. Optical transmission spectrum and XRD characterization both proved the high quality of the AlN template. The XRD (002) FWHM was about 53.7 arcsec and (102) FWHM was about 625 arcsec. The densities of screw TDs and edge TDs were estimated to be $\sim 6 \times 10^6$ cm^{-2} and $\sim 4.7 \times 10^9$ cm^{-2}. AlGaN of Al composition 80.2% was further grown on the AlN template. The RMS of the surface roughness was about 0.51 nm and clear steps could be seen. XRD reciprocal space mapping (RSM) was carried out to accurately determine the Al composition and relaxation status of the AlGaN epilayer. The XRD (002) rocking curve full width at half maximum (FWHM) was about 140 arcsec and (102) FWHM was about 537 arcsec. The density of screw TDs was estimated to be $\sim 4 \times 10^7$ cm^{-2} and that of edge TDs was $\sim 3.3 \times 10^9$ cm^{-2}. These values all prove the high quality of the AlN template and the AlGaN epilayer.

References

[1] Han J, Crawford M H, Shul R J, et al. AlGaN/GaN quantum well ultraviolet light emitting diodes. Appl Phys Lett, 1998, 73(12): 1688

[2] Nishida T, Kobayashi N. 346 nm emission from AlGaN multi-quantum-well light emitting diode. Physica Status Solidi A, 1999, 176(1): 45

[3] Kinoshita A, Hirayama H, Ainoya M, et al. Room-temperature operation at 333 nm of $Al_{0.03}Ga_{0.97}N/Al_{0.25}Ga_{0.75}N$ quantum-well light-emitting diodes with Mg-doped superlattice layers. Appl Phys Lett, 2000, 77(2): 175

[4] Khan M A, Shatalov M, Maruska H P, et al. III-nitride UV devices. Jpn J Appl Phys, 2005, 44(10): 7179

[5] Khan A, Balakrishnan K, Katona T. Ultraviolet light-emitting diodes based on group three nitrides. Nature Photonics, 2008, 2(2): 77

[6] Chichibu S F, Uedono A, Onuma T, et al. Origin of defect-insensitive emission probability in In-containing (Al,In,Ga)N alloy semiconductors. Nature Mater, 2006, 5(10): 810

[7] Morita D, Yamamoto M, Akaishi K, et al. Watt-class high-output-power 365 nm ultraviolet light-emitting diodes. Jpn J Appl Phys, 2004, 43(9A): 5945

[8] Zhang J, Hu X, Lunev A, et al. AlGaN deep-ultraviolet light-emitting diodes. Jpn J Appl Phys, 2005, 44(10): 7250

[9] Taniyasu Y, Kasu M, Makimoto T, et al. An aluminium nitride light-emitting diode with a wavelength of 210 nanometres. Nature, 2006, 441(7091): 325

[10] Kamiyama S, Iwaya M, Hayashi N, et al. Low-temperature-deposited AlGaN interlayer for improvement of AlGaN/GaN heterostructure. J Cryst Growth, 2001, 223(1/2): 83

[11] Iwaya M, Terao S, Hayashi N, et al. Realization of crack-free and high-quality thick $Al_xGa_{1-x}N$ for UV optoelectronics using low-temperature interlayer. Appl Surf Sci, 2000, 159: 405

[12] McAleese C, Kappers M J, Rayment F D G, et al. Strain effects of AlN interlayers for MOVPE growth of crack-free AlGaN and AlN/GaN multilayers on GaN. J Cryst Growth, 2004, 272(1–4): 475

[13] Khan M A, Skogman R A, van Hove J M, et al. Atomic layer epitaxy of GaN over sapphire using switched metalorganic chemical vapor deposition. Appl Phys Lett, 1992, 60(11): 1366

[14] Khan M A, Kuznia J N, Skogman R A, et al. Low pressure metalorganic chemical vapor deposition of AIN over sapphire substrates. Appl Phys Lett, 1992, 61(21): 2539

[15] Gay P, Hirsch P B, Kelly A. The estimation of dislocation densities in metals from X-ray data. Acta Metallurgica, 1953, 1(3): 315

[16] Lee S R, West A M, Allerman A A, et al. Effect of threading dislocations on the Bragg peakwidths of GaN, AlGaN, and AlN heterolayers. Appl Phys Lett, 2005, 86(24): 241904

[17] Nishida T, Akasaka T, Kobayashi N. Step-flow metalorganic vapor phase epitaxy of GaN on SiC substrates. Jpn J Appl Phys, 1998, 37(4B): L459

[18] Stephenson G B, Eastman J A, Thompson C, et al. Observation of growth modes during metal-organic chemical vapor deposition of GaN. Appl Phys Lett, 1999, 74(22): 3326

[19] Lorenz K, Gonsalves M, Kim W, et al. Comparative study of GaN and AlN nucleation layers and their role in growth of GaN on sapphire by metalorganic chemical vapor deposition. Appl Phys Lett, 2000, 77(21): 3391