JOURNALOF

Ceramic Processing Research

Heteroepitaxial growth of semipolar (11-22) GaN without low temperature GaN buffer layer grown on m-sapphire substrate

Sung-Nam Lee^{a,*}, Jihoon Kim^b and Hyunsoo Kim^c

^aOptoelectronic Materials & Devices Lab., Department of Nano-Optical Engineering, Korea Polytechnic University, Siheung 429-793, Republic of Korea

^bDevision of Advanced Materials Engineering, Kongju National University, Cheonan 331-717, Republic of Korea

^cSchool of Semiconductor and Chemical Engineering, Semiconductor Physics Research Center, Chonbuk National University, Jeonju 561-756, Republic of Korea

We investigated high quality semipolar (11-22) GaN epilayers were grown by novel 2-step growth method, which consisted of GaN (the first step) with N_2 atmosphere and GaN (the second step) with H_2 atmosphere at the growth temperature above 1000 °C without low temperature (LT) GaN or high temperature (HT) AlN buffer layer. As a nucleation layer, the thickness of GaN with N_2 atmosphere was varied from 0.3 to 0.1 µm. The full width at half maximum (FWHM) of X-ray ω -rocking curve (XRC) was decreased from 1088 to 951 arcsec with reducing thickness of nucleation layer. After fixing 0.1 µm-thick GaN with N_2 atmosphere, semipolar GaN epilayers were grown by controlling V/III, growth pressure and growth temperature under H_2 atmosphere. From these results, we could achieve the minimum XRC FWHM of 602 arcsec with increasing V/III, growth pressure, and growth temperature.

Key words: A1. High-resolution X-ray diffraction, A3. Metalorganic chemical vapor deposition, B1. Nitrides. PACS: 61.05.cp, 61.72.Uj, 68.55.ag, 78.67.De, 78.55.Cr.

Introduction

III-nitrides have been attractive wide bandgap semiconductor for optical and electronic device applications. In general, GaN-based devices are mainly grown on cplane GaN templates, which suffer from the quantum confinement Stark effect due to the existence of strong piezoelectric and spontaneous polarization [1-3]. The build-in electric fields along c-direction cause spatial separation of electron and hole that in turn gives rise to the restriction of carrier recombination efficiency, the reduction of oscillator strength, and the redshift of peak emission [1, 4]. To solve the limits of physical problems in III-nitrides, a lot of research groups have studied in the growth of nonpolar (11-20) a-plane and (10-10) mplane GaN-based devices for polarization free heterostructure [4-6]. Another approaches are (11-22), (10-1-1) and (10-1-3) semipolar III-nitrides devices [7-9]. It has been also found that semipolar (11-22) plane have higher In incorporation rate than nonpolar a- and mplane due to the crystallographic properties [10].

However, it is very difficult to obtain high quality semipolar (11-22) GaN grown on m-plane sapphire due to the significant anisotropic crystallographic difference [6, 8]. So far, most research groups have used conventional 2-step GaN growth approaches with LT-GaN or HT-AlN buffer to achieve high quality semipolar GaN [7-11]. However, the crystal and optical quality of semipolar GaN was still beginning stages. In this studywe introduced a high temperature (>1000 °C) GaN nucleation layer grown by only N₂ gas atmosphere instead of conventional LT-GaN or HT-AlN buffer layer. Based on novel N₂-GaN as a nucleation layer, we systematically investigated the high quality (11-22) semipolar GaN grown on m-plane sapphire by optimizing various growth parameters such as the thickness of N₂-GaN seed layer, growth temperature, V/III ratio, and growth pressure.

Experimental

High quality semipolar (11-22) GaN epilayers were directly grown on m-plane sapphires by using metalogranic chemical vapor deposition (MOCVD) system. Trimethylgallium (TMGa), ammonia (NH₃) were used as gallium, nitrogen sources, respectively. 2 µmthick semipolar GaN was grown by novel growth method, which consisted of GaN (the first step, N₂-GaN) with N₂ atmosphere and GaN (the second step, H₂-GaN) with H₂ atmosphere at the growth temperature above 1000 °C without LT-GaN or HT-AlN buffer layer [5, 12]. Before the growth of N₂-GaN layer, we carried out the nitridation process for m-sapphire substrate using NH₃ gas at 1050 °C for 5 min. And then, instead of LT-GaN buffer layer, the thickness of N₂-GaN as a

^{*}Corresponding author:

Tel:+82-31-8041-0721 Fax:+82-31-8041-0729

E-mail: snlee@kpu.ac.kr

s252



Fig. 1. The XRC FWHMs of semipolar (11-22) GaN/m-plane sapphire substrate as a function of thickness of N_2 -GaN as a nucleation layer.

seed GaN layer was varied from 100 to 300 nm. The growth temperature of N₂-GaN was 1030 °C. Subsequently, we grew H₂-GaN with different growth temperatures ($1030 \sim 1070$ °C), V/III ratios ($750 \sim 3000$), and growth pressures ($100 \sim 300$ torr) to obtain high crystal quality with the mirror-like surface morphology. The growth rate of semipolar (11-22) GaN was independent upon our growth conditions of growth temperatures and V/III ratios, while that of semipolar GaN was decreased from 1.0 to 0.85 nm/s with increasing the growth pressure.

To exclude the effect of film thickness on crystal quality and surface morphology, we fixed a constant film thickness of 2.0 μ m which was total thickness of N₂-GaN and H₂-GaN. The macroscopic surface morphologies of semipolar GaN templates were characterized by Nomarski optical microscope (NOM). From high resolution X-ray diffraction measurements, we investigated the crystal qualities of semipolar GaN grown by the different growth parameters. In particular, we choose the FWHMs of X-ray rocking curves (XRCs) with incident beam direction of [11-2-3], which represented minimum values of XRC FWHMs with different azimuth angles from -90 to 90 °.

Results and discussion

In the growth of GaN/sapphire, at the initial stage many island-shaped nuclei are formed on the substrate, which coalesce with a three-dimensional growth mode. With further growth, the large-scale pits are buried and the surface gradually becomes planarization [13]. In this growth procedure, initial stage is very important to affect further growth mode. The desorption of nitrogen from the GaN surface can be suppressed by using the N₂ atmosphere [15]. Because of this, we first investigated the influence of N₂-GaN seed GaN layer instead of LT-GaN buffer layer on surface morphology and crystal quality of semipolar GaN. Fig. 1 showed XRCs FWHMs of semipolar GaN as a function of



Fig. 2. The XRC FWHMs of semipolar (11-22) GaN/m-plane sapphire substrate as a function of growth temperature of H_2 -GaN.

thickness of N₂-GaN seed layer. Figs. 5 (a-c) were NOM images of semipolar GaN epilayer with different thickness of N2-GaN layer. No surface pits were observed on all samples and relative smooth surface morphologies were obtained by our 2-step growth technique. However, in spite of obtaining the smooth surface with pit-free, surface morphology was more undulated by increasing the thickness of N₂-GaN. It implied that N2-GaN layer could play an important role in nucleation layer instead of an LT-GaN buffer layer to achieve the high quality semipolar GaN. In particular, thinner N2-GaN formed more mirror-like surface structure than thicker N2-GaN epilayer. In addition, the XRC FWHMs was decreased from 1088 to 951 arcsec with reducing the thickness of N2-GaN layer. It indicated that thinner N2-GaN could also improve crystal quality as well as surface morphology. In general, the nitrogen atmospheric growth in MOCVD system would accelerate the density and size of nucleation layer due to the increase of adatom diffusion length [15]. We believed that thicker (> 100 nm) N_2 -GaN would play a role in the increase of the density and the size of nucleation layer as a buffer layer, resulting in the rough surface and the high mosaicity of semipolar (11-22) GaN epilayer due to the large size and the density of seed layer on the same an LT-GaN buffer layer as c-plane GaN/sapphire. From these results, we would guess that thicker N2-GaN with relative poor crystal quality would significantly affect the crystal quality of H₂-GaN and should be limited within 0.1 µmthick N₂-GaN in our 2-step growth technique.

We have also studied the effect of H_2 -GaN growth temperature on crystal qualities and surface morphology of semipolar GaN with a constant 100 nm-thick N₂-GaN as shown in Fig. 2. The surface morphology was not significantly degraded but slightly roughed by increasing growth temperature as shown in Figs. 5 (d-f). It is believed that in spite of increasing lateral to vertical growth rate at a high growth temperature, the surface atoms had a tendency to be desorbed at higher growth







Fig. 4. The XRC FWHMs of semipolar (11-22) GaN grown with different pressure of H₂-GaN.



Fig. 5. The Normalski optical microscope images of semipolar GaN with different N_2 -GaN thickness (a-c), the growth temperature of H_2 -GaN layers (d-f), semipolar GaN with V/III ratios (g-i) and H_2 -GaN growth pressure (j-l).

temperature, resulting in the deterioration of surface structure. However, the FWHM values of XRCs were decreased from 1003 to 890 arcsec by increasing the growth temperature. It indicated that crystal quality of semipolar GaN was enhanced by increasing the growth temperature. We guessed that the reduction of crystal defects may be caused by a predominant coalescence step due to the increase of lateral growth rate at a high temperature.

Comparing with the V/III ratio of conventional c-

plane GaN, low V/III ratio had been used to obtain the smooth surface morphology for semipolar GaN epilayer. In this study, we reduced V/III ratio from 3000 to 750 with increasing the flow rate of TMGa. Fig. 3 showed the XRC FWHMs of semipolar GaN with 50 nm-thick N₂-GaN as a function of V/III ratio of H₂-GaN layer grown at 1030 °C. Figs. 5 (g-i) are the NOM images of each sample grown at different growth temperatures. XRC FWHMs were significantly decreased from 1309 to 758 arcsec with increasing V/III ratio, while best surface morphology was obtained at the V/ III of 1500. From crystallinity of view, low V/III ratio may generate crystal defects due to the deficiency of N atoms. In addition, it is known that low V/III ratio could increase the growth rate for c-direction, which would develop many arrowhead-like surface structures.

The growth pressure was one of most important parameters in MOCVD system, which could modify the growth mode due to the change of gas flow dynamics [14]. To study the effect of growth pressure, we investigated XRC FWHMs and surface morphologies of semipolar GaN grown by different growth pressures of H₂-GaN layer as show in Fig. 4. XRC FWHMs was significantly reduced from 990 to 602 arcsec with increasing the growth pressure, which is one of best results in the heteroepitaxial growth of semipolar (11-22) GaN with smooth surface morphology. It showed that the crystal quality of semipolar GaN could be significantly by increasing the growth pressure. However, the surface morphology of semipolar GaN was mirror-like at the pressure of 100 torr, while that of semipolar GaN grown at the pressure of 300 torr was rough surface with undulated structures as shown in Figs. 5 (j-l).

Conclusions

We studied the effects of thickness of N_2 -GaN layer and growth temperature, V/III ratio, and pressure on the crystal quality and surface morphology of semipolar (11-22) GaN grown on m-plane sapphire. Instead of LT-GaN buffer layer as a nucleation layer, N_2 -GaN layer could effectively developed to obtain the high quality semipolar GaN. The crystal quality and surface morphology was significantly enhanced by reducing the thickness of N_2 -GaN layer. Additionally, the crystal quality of semipolar GaN was improved by increasing growth temperature, V/III ratio, and growth pressure, while the surface morphology was enhanced by reducing the V/III ratio and growth pressure. From these results, we could obtain the narrowest FWHM of 602 arcsec for semipolar (11-22) GaN/m-sapphire using novel 2-step growth with a N_2 -GaN nucleation layer.

Acknowledgement

This research was supported by the IT R&D program of MKE/KEIT [10039151, The development of 200 mW level high power green (525 nm) LED for full color display] and Core Corporation Research Program (2010-0026523) through the National Research Foundation of Korea (NRF) funded by the Ministry of Education, Science and Technology.

References

- P. Waltereit, O. Brandt, A. Trampert, H.T. Grahn, J. Menniger, M. Ramsteiner, M. Reiche, and K.H. Ploog, Nature 406 (2000) 865.
- S.F. Chichibu, A.C. Abare, M.S. Minsky, S. Keller, S.B. Fleischer, J.E. Bowers, E. Hu, U.K. Mishra, L.A. Coldren, S.P. DenBaars, and T. Sota, Appl. Phys. Lett. 73 (1998) 2006.
- T. Takeuchi, S. Sota, M. Katsuragawa, M. Komori, H. Takeuchi, H. Amano, and I. Akasaki, Jpn. J. Appl. Phys. 36 (1997) L382.
- M.D. Craven, S.H. Lim, F. Wu, J.S. Speck, and S.P. DenBaars, Appl. Phys. Lett. 81(2002) 469.
- S.N. Lee, H.S. Paek, J.K. Son, T. Sakong, O.H. Nam, and Y. Park, J. Cryst. Growth. 307 (2007) 368.
- S.N. Lee, H.S. Paek, H. Kim, Y.M. Park, T. Jang, and Y. Park, Appl. Phys. Lett., 92 (2008) 111106.
- A. Tyagi, H. Zhong, R.B. Chung, D.F. Feezell, M. Saito, K. Fujito, J.S. Speck, S.P. Denbaars, and S. Nakamura, J. Jpn. Appl. Phys. 46 (2007) L444.
- M.J. Kappers, J.L. Hollander, C. McAleese, C.F. Johnston, R.F. Broom, J.S. Barnard, M.E. Vickers, C.J. Humphreys, J. Cryst. Growth 300 (2007) 155.
- R. Sharma, P.M. Pattison, H. Masu, R.M. Farrell, T.J. Baker, B.A. Haskell, F. Wu, S.P. DenBaars, J.S. Speck, and S. Nakamura, Appl. Phys. Lett. 87 (2005) 231110.
- Y. Kawakami, K. Nishizuka, D. Yamada, A. Kaneta, M. Funato, Y. Narukawa, and T. Mukai, Appl. Phys. Lett. 90(2007) 261912.
- Q. Sun, B. Leung, C.D. Yerino, Y. Zhang, J. Han, Appl. Phys. Lett. 95(2009) 231904.
- S.N. Lee, K.K. Kim, O.H. Nam, J.H. Kim, and H. Kim, Phys. Stat. Solidi. C, Phys. Stat. Solidi. C, V7(2010) 2043.
- I. Akasaki, H. Amano, Y. Koide, K. Hiramatsu and N. Sawai. J. Cryst. Growth 98 (1989) 209.
- R. Miyagawa, M. Narukawa, B. Ma, H. Miyake, K. Hiramatsu, J. Cryst. Growth 310 (2008) 4979.
- Y. Kobayashi and N. Kobayashi, J. Cryst. Growth. 189/190 (1998) 301.