Applied Surface Science 257 (2011) 2415–2418

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Review

Nonpolar a-plane GaN grown on r-plane sapphire using multilayer AlN buffer by metalorganic chemical vapor deposition

C.H. Chiang[a,*], K.M. Chen[a], Y.H. Wu[a], Y.S. Yeh[a], W.I. Lee[a], J.F. Chen[a], K.L. Lin[b], Y.L. Hsiao[b], W.C. Huang[b], E.Y. Chang[b]

[a] Department of Electrophysics, National Chiao Tung University, Hsinchu, Taiwan, ROC
[b] Department of Materials Science and Engineering, National Chiao Tung University, Hsinchu, Taiwan, ROC

ARTICLE INFO

Article history:
Received 5 January 2010
Received in revised form 6 October 2010
Accepted 14 October 2010
Available online 20 October 2010

Keywords:
GaN
Crystal morphology
Nonpolar
MOCVD

ABSTRACT

Mirror-like and pit-free non-polar a-plane (1 1 −2 0) GaN films are grown on r-plane (1 −1 0 2) sapphire substrates using metalorganic chemical vapor deposition (MOCVD) with multilayer high-low-high temperature AlN buffer layers. The buffer layer structure and film quality are essential to the growth of a flat, crack-free and pit-free a-plane GaN film. The multilayer AlN buffer structure includes a thin low-temperature-deposited AlN (LT-AlN) layer inserted into the high-temperature-deposited AlN (HT-AlN) layer. The results demonstrate that the multilayer AlN buffer structure can improve the surface morphology of the upper a-plane GaN film. The grown multilayer AlN buffer structure reduced the tensile stress on the AlN buffer layers and increased the compressive stress on the a-plane GaN film. The multilayer AlN buffer structure markedly improves the surface morphology of the a-plane GaN film, as revealed by scanning electron microscopy. The effects of various growth V/III ratios was investigated to obtain a-plane GaN films with better surface morphology. The mean roughness of the surface was 1.02 nm, as revealed by atomic force microscopy. Accordingly, the multilayer AlN buffer structure improves the surface morphology and facilitates the complete coalescence of the a-plane GaN layer.

Contents

1. Introduction 2415
2. Experimental 2416
3. Results and discussion 2417
4. Conclusions 2418
Acknowledgments 2418
References 2418

1. Introduction

Wurtzite III-nitrides are of particular interest in scientific research and practical applications. They have a wide range of applications in visible or ultraviolet light-emitting diodes and laser diodes grown on c-plane sapphires, r-plane sapphires, SiC or free-standing substrates. Currently, most nitride-base devices are fabricated with a c-plane [0 0 0 1] orientation, including InGaN/GaN strained-layer quantum well (QW) structures. A larger lattice mismatch between GaN and InGaN makes the epitaxial layers highly strained. Accordingly, strong electrostatic fields form parallel to the growth direction, causing the electrons and holes to be spatially separated in the QW [1–3]. Such separation reduces the recombination ratio for emission and increases the radiative lifetime at the expense of the quantum efficiency and the emission wavelength [4]. This phenomenon is called the quantum-confined stark effect (QCSE). To solve these problems, various approaches have been proposed to grow non-polar or semi-polar GaN to eliminate polarization effects, For example, a study of optimal growth conditions has demonstrated that a lower reactor pressure, a higher growth temperature, and a lower V/III ratio critically promote the growth of a high-quality and pit-free a-plane GaN [5], by the flow-rate modulation epitaxy technique [6], and that the use of the lateral epitaxial overgrowth (LEO) [7] or sidewall lateral epitaxial overgrowth (SLEO) [8] technique can improve an a-plane GaN on r-plane sapphire substrate by reducing the density of threading disloca-

---

[*] Corresponding author.
E-mail address: ziglar0316@gmail.com (C.H. Chiang).

0169-4332/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.apsusc.2010.10.059

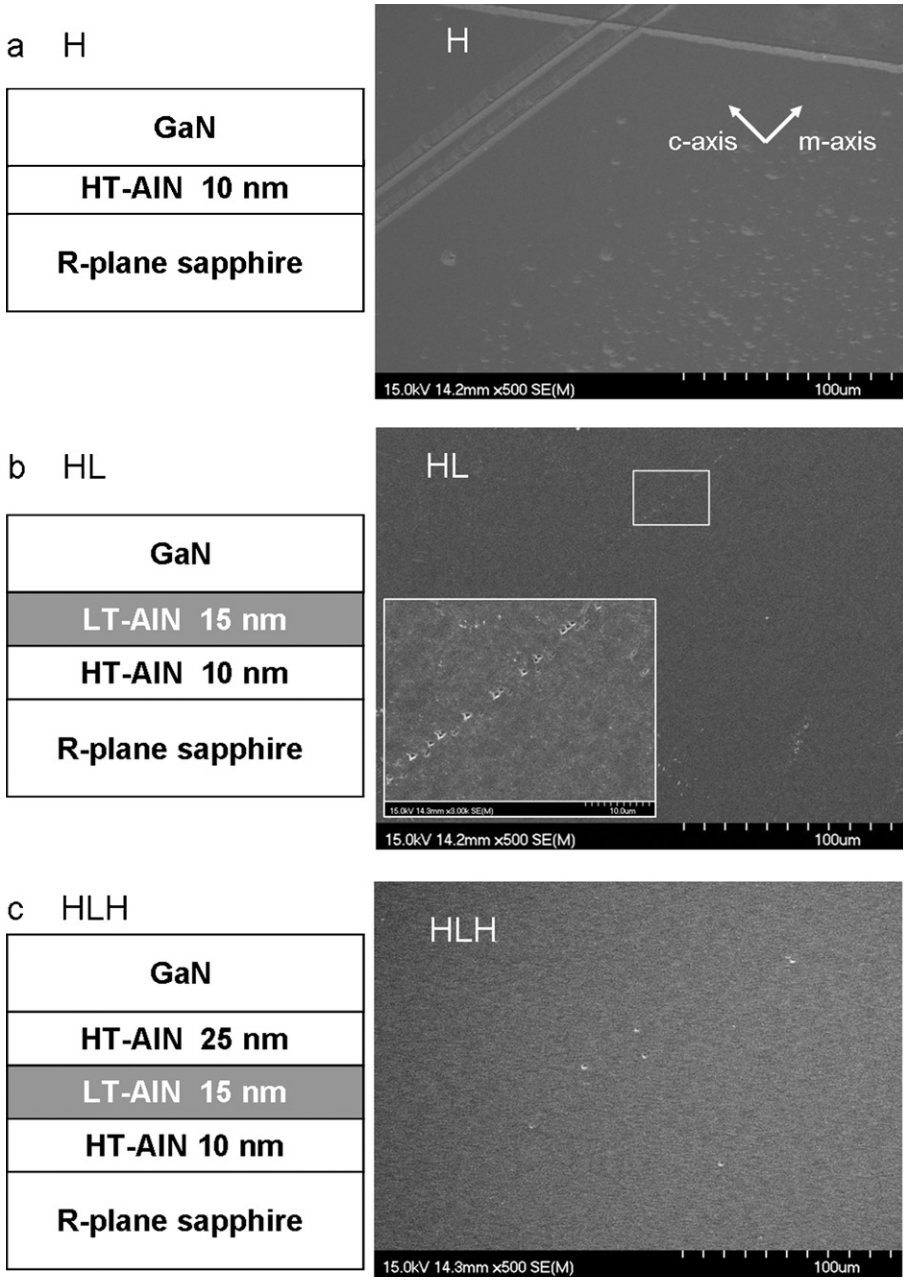

Fig. 1. SEM images and cross-sectional structural schematic diagrams of a-plane GaN grown on r-plane sapphire with various buffer layers. (a) HT-AlN buffer layer. (b) LT-AlN/HT-AlN buffer layer. (c) Sandwiched AlN buffer layer. The inset magnifies the area enclosed by the rectangular frame in SEM image (b).

tions. The r-plane sapphire has attracted much interest due to its low cost as compared with other substrates such as $LiAlO_2$ and SiC. The important issues around a-plane GaN on r-plane sapphire substrates are the lattice constant mismatch, the large thermal expansion coefficients and the higher growth rate in the c-direction than in the m-direction. These results reveal an a-plane GaN stripe along the c-direction. This investigation presents the improvement of crystalline quality of a pit-free a-plane GaN on r-plane sapphire substrate using the multilayer AlN buffer structure. An intermediate temperature interlayer can be used to grow high-quality GaN on c-plane sapphire substrate and thick crack-free GaN on the Si substrate. The intermediate temperature interlayer technique can markedly reduce the density of threading dislocations when GaN is grown on c-plane sapphire [9,10]. Also, an low-temperature-deposited AlN (LT-AlN) interlayer reduces the stress between a GaN layer and Si substrate [11]. GaN grown with an inserted LT-AlN interlayer exhibit suppressed tensile stress [12]. However, the effects of the buffer layer structure and growth parameters on the quality of a grown non-polar a-plane GaN film has not yet been studied.

2. Experimental

All samples in this study are grown on (1 −1 0 2) r-sapphire using a low-pressure metalorganic chemical vapor deposition (LP-MOCVD) reactor(EMCORE D-180). The gallium, aluminum and nitrogen sources are trimethylgallium (TMGa), trimethyaluminum (TMAl) and ammonia (NH_3), respectively. The multilayer AlN buffer structure was used instead of conventional AlN layers. The epilayer structure of all samples comprised, from bottom to top, a high-temperature-deposited AlN (HT-AlN) nucleation layer grown

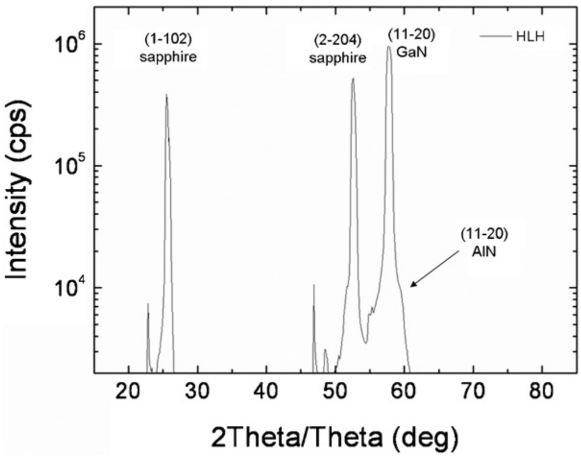

Fig. 2. $2\theta-\omega$ scan of a-plane GaN grown with sandwiched AlN buffer layer on r-plane sapphire.

at 1100 °C with thickness of 20 nm, a LT-AlN nucleation layer grown at 800 °C with thickness of 30 nm, and an HT-AlN nucleation layer grown at 1100 °C with thickness of 50 nm. The growth pressure and V/III ratio of the nucleation layers were 100 Torr and 1648, respectively. Then, the 2.5 µm-thick undoped a-GaN layer was grown at 1100 °C, a reactor pressure of 100 Torr, and with H_2 carrier gas. In this work, the surface morphology of pit-free a-GaN on r-sapphire substrate was improved using multilayer AlN buffer structure. To evaluate suitable parameters for the growth of a-GaN on multilayer AlN buffer, the effect of TMGa flow-rate (V/III ratio of 1216, 912, and 729) was studied. The surface morphology and microstructure of the samples were investigated by scanning electronic microscopy (SEM) and atomic force microscopy (AFM), respectively. The growth process was observed by the *in situ* monitoring of optical reflectance.

3. Results and discussion

This work presents the improvement in the surface morphology of the pit-free a-GaN on r-sapphire substrate using multilayer AlN buffer structure. The buffer layer between a-GaN and r-sapphire substrate is important to the high-quality a-GaN films on r-sapphire. On the other hand, the value of the thermal expansion coefficients of AlN is between GaN and Si, which can also be use to reduce the thermal mismatch between a-GaN and r-sapphire substrate. Fig. 1(a–c) presents the schematic structure and SEM images of the a-GaN with layers a-GaN/HT-AlN/r-sapphire, a-GaN/LT-AlN/HT-AlN/r-sapphire and a-GaN/HT-AlN/LT-AlN/HT-AlN/r-sapphire, [denoted H, LH, and HLH,] respectively. The single layer AlN buffer, random pits with a high density can be seen along with cracks. The sample LH can eliminate cracks and reduce the pit density, the sample HLH further reduce it to nearly smooth surface. The cracks were observed in the m-direction and the pit density was reduced around the corner of the cracked a-GaN, as shown in Fig. 1(a). The results suggest that lattice mismatch reduced near cracked a-GaN film. Therefore, a large lattice mismatch between a-GaN and r-sapphire produces cracks and a high pit density on the film grown, the pit density and creak density are approximately $8 \times 10^6\,\mathrm{cm}^{-2}$ with pits of size 1–6 µm and $3\,\mathrm{mm}^{-1}$. The sample LH can eliminate cracks and reduce the pit density from $8 \times 10^6\,\mathrm{cm}^{-2}$ to $3.5 \times 10^5\,\mathrm{cm}^{-2}$. Fig. 1(b) indicates that the pits begin to coalesce along the m-axis of sample LH. This behavior appears to be consistent with that of sample H in the SEM image in Fig. 1(a), indicating that the cracks formed in the m-direction by the coalescence of pits. The Inset SEM image shows that the corresponding to the area enclosed by the white box. The result revealed the pits aligned along the m-axis (perpendicular to the c-axis). This finding is similarly to that in the literature [13]. Therefore, the buffer layer structure and film quality are essential to the growth of a flat, crack-free and pit-free a-GaN film. Fig. 1(c) shows SEM images of a nearly smooth surface by the multilayer AlN buffer structure. The pits clearly become smaller and their density on the surface decreases. This result can be explained by the insertion of a thin LT-AlN layer into the HT-AlN layer, which reduces the stress between the a-GaN film and the r-sapphire substrate. However, many defects and cracks formed on the first HT-AlN layer because of the relaxation of the stress in this layer. The second layer of the LT-AlN film was used as the nucleation layer of the growth of the top HT-AlN layer and also prevented the propagation of cracks and defects from the first HT-AlN layer to the top HT-AlN layer. The LT-AlN interlayer effectively blocks the propagating defects in the growth direction and reduces the dislocation density [10]. The third layer of the HT-AlN film forms a high-quality AlN with a lower defect density to pro-

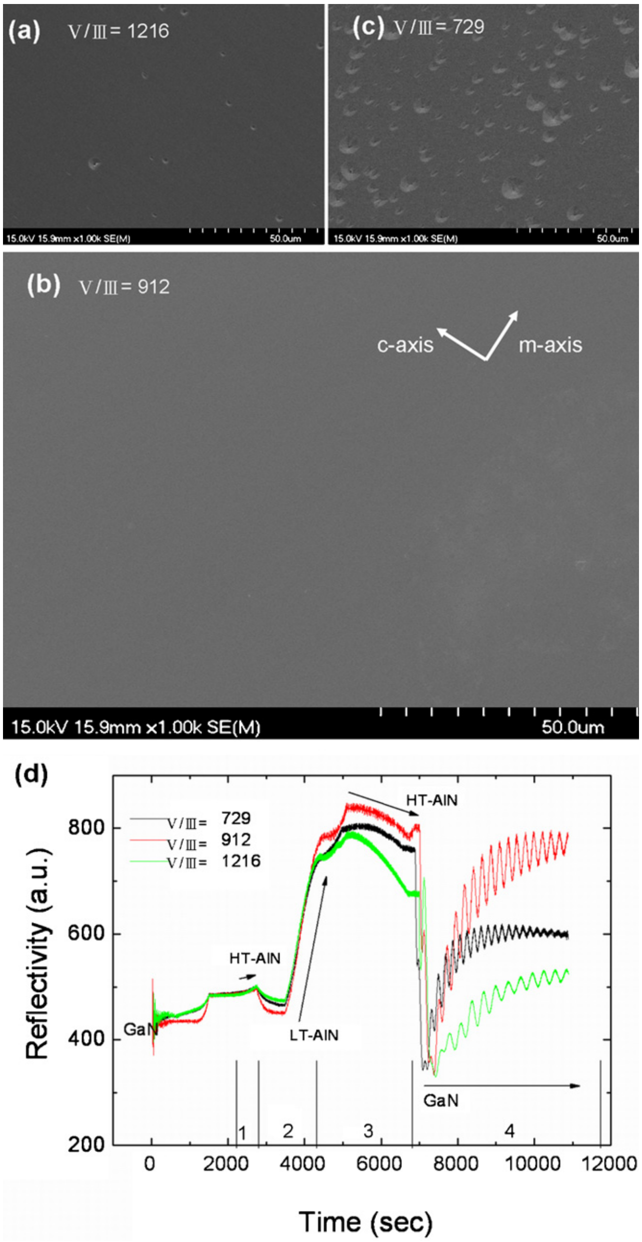

Fig. 3. SEM images (a) and traces of *in situ* optical reflectivity measurements (b) of the whole growth process of a-plane GaN film on sandwiched AlN buffer layer at 100 Torr with V/III ratios of 1216, 912 and 729.

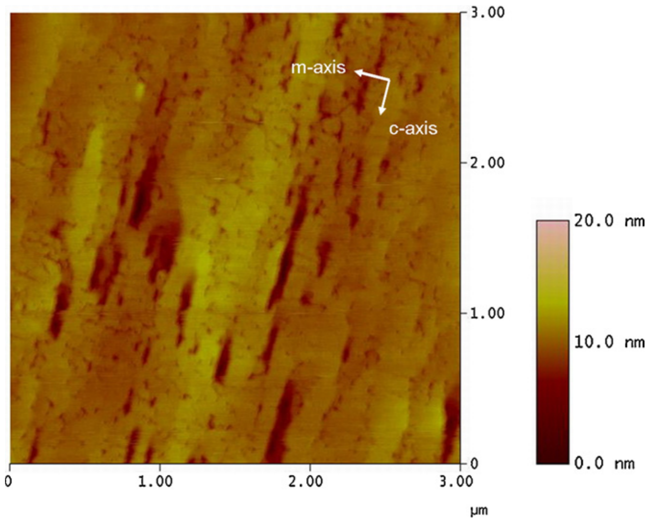

Fig. 4. AFM image of a-plane GaN grown under 100 Torr at V/III ratio of 912.

mote the further growth of the a-GaN film. Hence, the growth a-GaN with the multilayer AlN buffer structure on r-sapphire markedly improves the surface morphology of the a-GaN film. X-ray diffraction measurements of the HLH sample with a $2\theta-\omega$ scan yielded sapphire (1 −1 0 2), (2 −2 0 4), AlN (1 1 −2 0), and GaN (1 1 −2 0) reflections, as shown in Fig. 2, indicating that the surface of the GaN film is a-plane with multilayer AlN buffer structure. To optimize further a-GaN films with the multilayer AlN buffer structure, the influence of the TMGa flow-rate was studied. The three samples shown here have the same structure, with different morphology on the surface. The V/III ratio was adjusted from 1216 to 912 and 729. The surface morphologies of all samples were observed by SEM and the results shown in Fig. 3(a–c). Fig. 3(b) is the result of the sample grown at 1100 °C and at 100 Torr with multilayer AlN buffer structure. The pit-free a-GaN is observed at a V/III ratio of 912, revealed by the SEM image at 1000 × magnification. Further decreasing flow-rate of TMGa, the density of small-size pits were increased ($\sim 2.4 \times 10^7$ cm^{-2}), as shown in Fig. 3(c). The further decrease in the TMGa flow-rate could cause some N atoms not to react with Ga toms, which degraded the surface morphology. Five traces are divided into four regions, which correspond to the four growth stages of a-GaN grown on the multilayer AlN buffer structure, as follows; (1) HT-AlN buffer layer deposition, (2) LT-AlN buffer layer deposition, (3) HT-AlN buffer layer deposition, (4) growth of a-GaN film. The surface of the a-GaN film becomes smooth and the intensity of the *in situ* optical reflectivity increases. The reflectivity intensity of the a-GaN with a V/III ratio of 912 markedly exceeds that of the other samples. These results show that, with lower TMGa flow-rate, the surface morphology of a-plane GaN can achieve pit-free and smooth due to the slower growth ratio. The oscillation of the reflectivity intensity indicates the two-dimensional growth of the a-GaN layer, as shown in Fig. 3(d). The frequency of oscillation of the reflectivity intensity represents the growth rate. Hence, the oscillation frequency of a-GaN grown on the multilayer AlN buffer structure is increased as the flow-rate of TMGa increase. Fig. 4 shows a representative $3 \times 3\,\mu m^2$ AFM image of a-GaN grown at 100 Torr and at a V/III ratio of 912 with the multilayer AlN buffer structure. The AFM root mean square (RMS) roughness was about 1.02 nm. Insertion of a thin LT-AlN layer into the HT-AlN layer effectively reduces strain improving the surface morphology of a-GaN.

4. Conclusions

This study investigated the effects of how the structure of the buffer layers and the TMGa flow-rate affects the surface morphology of a-plane GaN grown on r-plane sapphire. The multilayer AlN buffer structure was adopted to improve the surface morphology and facilitate the complete coalescence of the a-plane GaN layer. The mean roughness of the surface morphology was 1.02 nm. The multilayer AlN buffer structure markedly improved the surface morphology of the a-plane GaN film in the SEM images. The use of the multilayer AlN buffer structure suppressed the tensile stress on the AlN film and increased the compressive stress of the grown a-plane GaN film. The unique multilayer AlN buffer structure provides an effective and convenient means of improving the crystalline quality of a-plane GaN and fabricated non-polar devices.

Acknowledgments

The authors would like to thank the National Science Council of the Republic of China, Taiwan (Contract No. NSC 96-2112-M-009 -034 -MY3), and the Ministry of Education under the ATU program of for financially supporting this research.

References

[1] T. Takeuchi, C. Wetzel, S. Yamaguchi, H. Sakai, H. Amano, I. Akasaki, Y. Kaneko, S. Nakagawa, Y. Yamaoka, N. Yamada, Appl. Phys. Lett. 73 (1998) 1691.
[2] P. Lefebvre, A. Morel, M. Gallart, T. Taliercio, J. Allegre, B. Gil, H. Mathieu, B. Damilano, N. Grandjean, J. Massies, Appl. Phys. Lett. 78 (2001) 1252.
[3] J.S. Im, H. Kollmer, J. Off, A. Sohmer, F. Scholz, A. Hangleiter, Phys. Rev. B 57 (1998) R9435.
[4] P. Waltereit, O. Brandt, A. Trampert, H.T. Grahn, J. Menniger, M. Ramsteiner, M. Reiche, K.H. Ploog, Nature 406 (2000) 865.
[5] X. Ni, Y. Fu, Y.T. Moon, N. Biyikli, H. Morkoc, J. Cryst. Growth 290 (2006) 166.
[6] J.J. Huang, T.Y. Tang, C.F. Huang, C.C. Yang, J. Cryst. Growth 310 (2008) 2712.
[7] M.D. Craven, S.H. Lim, F. Wu, J.S. Speck, S.P. DenBaars, Appl. Phys. Lett. 81 (2002) 1201.
[8] B.M. Imer, F. Wu, S.P. DenBaars, J.S. Speck, Appl. Phys. Lett. 88 (2006) 061908.
[9] M. Iwaya, T. Takeuchi, S. Yamaguchi, C. Wetzel, H. Amano, I. Akasaki, Jpn. J. Appl. Phys. 37 (1998) L316.
[10] E.D. Bourret-Courchesne, S. Kellermann, K.M. Yu, M. Benamara, Z. Liliental-Weber, J. Washburn, S.J.C. Irvine, A. Stafford, Appl. Phys. Lett. 77 (2000) 3562.
[11] J. Blasing, A. Reiher, A. Dadgar, A. Diez, A. Krost, Appl. Phys. Lett. 81 (2002) 2722.
[12] H. Amano, M. Iwaya, T. Kashima, M. Katsuragawa, I. Akasaki, J. Han, S. Hearne, J.A. Floro, E. Chason, J. Figiel, Jpn. J. Appl. Phys. 37 (1998) L1540.
[13] M.D. Craven, F. Wu, A. Chakraborty, B. Imer, U.K. Mishra, S.P. DenBaars, J.S. Speck, Appl. Phys. Lett. 84 (2004) 1281.