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Nitridation effect of the α-Al_2O_3 substrates on the quality of the GaN films grown by pulsed laser deposition

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Abstract: GaN films have been grown on the nitrided and non-nitrided α-Al_2O_3 substrates by pulsed laser deposition (PLD). The surface morphologies and structural properties of these as-grown GaN films have been investigated carefully. It reveals that, when the nitridation process is performed on the α-Al_2O_3 substrates, the surface morphologies and structural properties of the as-grown GaN films grown on the as-nitrided α-Al_2O_3 substrates are improved dramatically. The effect of nitridation on the properties of GaN films and the growth mechanism of GaN films on nitrided α-Al_2O_3 substrates by PLD have been carefully studied. An effective approach to achieve high-quality GaN films on α-Al_2O_3 substrates by PLD is hence presented.

Keywords: α-Al_2O_3 substrates; pulsed laser deposition; nitridation; effect; mechanism.

1. Introduction

Nowadays, GaN and its related III-nitrides have generated a great deal of interest due to their superior properties, which make them possible for the application in light-emitting diodes, laser diodes, field effect transistors, photo-detectors, *etc*.[1-5] As usual, GaN films are grown by metal-organic chemical vapor deposition (MOCVD).[6-8] However, it requires a high growth temperature, which means the high consumptions of both power and cost.[9-10] Meanwhile, toxic metal-organic chemicals are often employed as the growth precursors in MOCVD chamber. This

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may inevitably cause environmental pollution.[11-12] To solve this inherent problem, pulsed laser deposition (PLD) technique has been introduced to grow III-nitrides recently.[13-14] In comparison with MOCVD, PLD employs a highly energetic laser to ablate target, which can ensure the species arrive at the substrates with highly kinetic energy.[15-16] In this case, it can realize the epitaxial growth of GaN at a relatively low growth temperature. Meanwhile, the sources for PLD growth are targets and nitrogen/oxygen, so the pollution produced by PLD is much less.[17-18] Furthermore, the large-size thin films with homogeneous thickness distribution have been obtained with the laser rastering technology integrated in PLD.[16-19] These advantages have therefore raised concerns on the study of PLD.

Previous works have studied on the growth of GaN films on α-Al_2O_3 substrates by PLD.[20-21] It is reported that by employing the procedure of nitridation, epitaxial GaN films have been grown by PLD on nitrided α-Al_2O_3 substrates successfully.[21-23] However, the effect of α-Al_2O_3 substrates nitridation on the properties of GaN films during PLD growth, as well as the growth mechanism of GaN on nitrided α-Al_2O_3 substrates by PLD, lacks thorough study.[24-27]

In this work, we investigate on the effect of α-Al_2O_3 substrates nitridation on the properties of GaN films grown by PLD systematically, and propose the growth mechanism of GaN films on nitrided α-Al_2O_3 substrates by PLD. An effective approach to achieve high-quality GaN films on α-Al_2O_3 substrates by PLD is hence presented.

2. Experimental

The α-Al_2O_3 substrates were firstly put into a ultra-high vacuum (UHV) load-lock chamber with a background pressure of 1.0×10^{-8} Torr, and then were degassed at 200 ºC for 30 min. Afterwards, they were transferred into the UHV growth chamber with a background pressure of 1.0×10^{-10} Torr, followed by a 60 min annealing at 850 ºC to remove the surface contaminations. Before the epitaxial growth, substrates were nitrided at 850 ºC for 60 min under the pressure of 4 mTorr in a high-purity nitrogen (7N) plasma ambient produced by a radio-frequency plasma generator attached to the PLD system and working at 500 W. Subsequently, a KrF excimer laser light (λ=248 nm, t=20 ns) was used to ablate the high-purity Ga (7N) in a nitrogen plasma ambient at pressure of 10 mTorr. The energy density of the laser was set at 3.0 $J.cm^{-2}$ with a pulse repetition of 30 Hz. The ablated species were then directed onto the substrates, which were mounted 5 cm away from

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the target and were maintained at 750 ºC for the growth of GaN for 10-60 min. In order to investigate the effect of α-Al_2O_3 substrates nitridation on the properties of GaN films, another epitaxial growth was conducted under the same growth conditions except for the non-nitridation of α-Al_2O_3 substrates. The schematic diagram for PLD growth of GaN films on α-Al_2O_3 substrates is shown in Fig. 1. The as-grown GaN films were characterized by *in-situ* reflection high energy electron diffraction (RHEED), field emission scanning electron microscopy (FESEM, Nova Nano SEM 430 Holland), atomic force microscopy (AFM, MFP-3D-S Asylum, American), high-resolution X-ray diffraction (HRXRD, Bruker D8 X-ray diffractometer with Cu Kα1 X-ray source λ=1.5406 Å) and high-resolution transmission electron microscopy (HRTEM, JEOL 3000F, field emission gun TEM working at a voltage of 300 kV, which gives a point to point resolution of 0.17 nm).

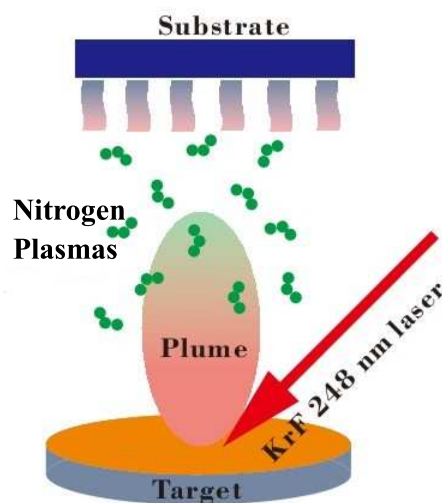

Fig. 1. Schematic diagram for PLD growth of GaN films on α-Al_2O_3 substrates.

3. Results and discussion

In-situ RHEED is used to monitor the GaN films grown on α-Al_2O_3 substrates during the growth. Fig. 2a is RHEED patterns for α-Al_2O_3 substrates before the annealing process. One can hardly observe the sharp patterns. After the 60-minute annealing process at 750 ºC, sharply spotty RHEED patterns along the α-Al_2O_3 [1-100] direction can be clearly identified, as shown in Fig. 2b. Clearer and sharper RHEED can be found after the nitridation process, as shown in Fig. 2c, which agrees well with the RHEED patterns for wurtzite-structured AlN films along the direction of

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[11-20]. In this case, the AlN thin films with very flat surfaces are obtained. After the epitaxial growth of ~300 nm-thick GaN films on nitrided α-Al_2O_3 substrates, one can clearly find the streaky RHEED patterns along the GaN[11-20] direction, which reveals that GaN films with smooth and flat surface have been grown, as shown in Fig. 2d. Conclusively, these results demonstrate that single-crystalline GaN films have been grown on nitrided α-Al_2O_3 substrates with an in-plane epitaxial relationship of GaN[11-20]//α-Al_2O_3[1-100].[28-29]

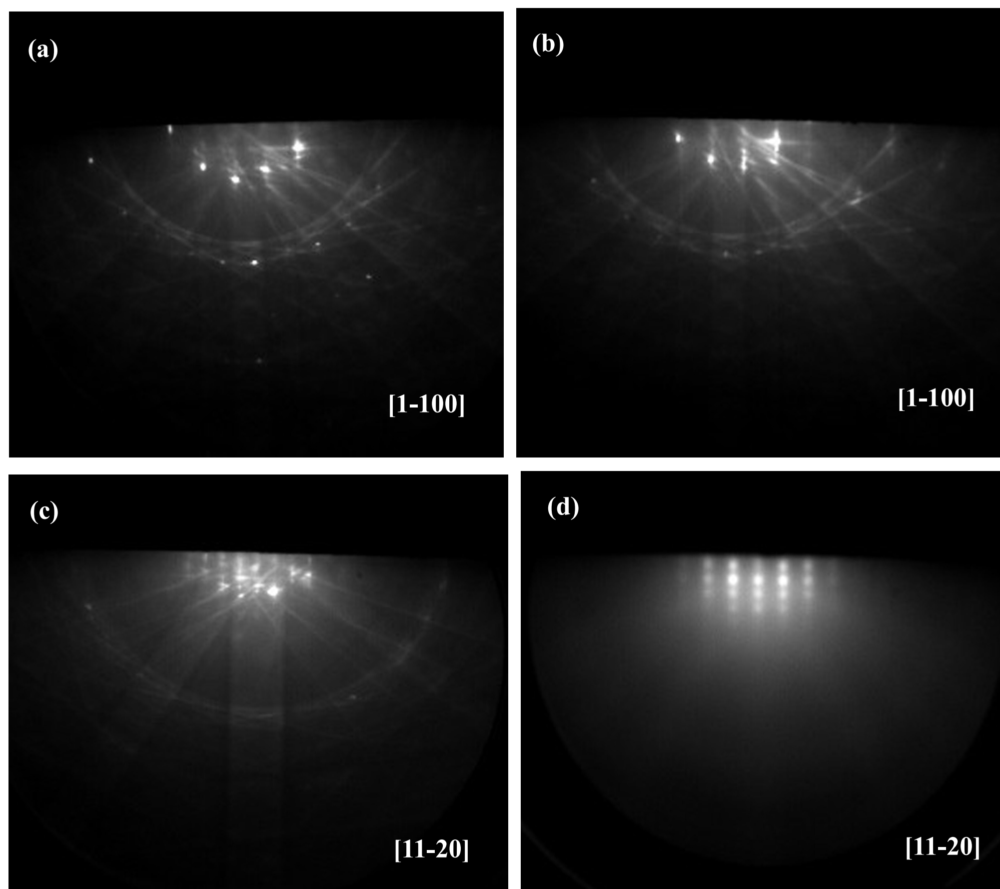

Fig. 2. RHEED patterns for α-Al_2O_3 substrates (a) before and (b) after annealing process at 750 ºC for 60 min, and (c) after nitridation process at 750 ºC. (d) RHEED patterns for ~300 nm-thick GaN films grown at 750 ºC.

The surface morphologies for GaN films grown on α-Al_2O_3 substrates are studied by FESEM and AFM, respectively. Fig. 3a is a FESEM image for the ~300 nm-thick GaN films grown on non-nitrided α-Al_2O_3 substrates at 750 ºC, from which one can clearly identify the particulates and islands distributed on the GaN surface with the root-mean-square (RMS) roughness of 3.1 nm,

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indicating the very rough surface. If the ~300 nm-thick GaN films are grown on the nitrided α-Al_2O_3 substrates at 750 ºC, which reveal the very smooth and flat surface with the RMS roughness of 1.2 nm measured by AFM, as shown in Fig. 3b. The possible reason for this difference is the nitridation process. The nitridation process leads to the formation of AlN layer, enhances the migration of GaN precursors on the surface, and thereby flats the GaN surface. Furthermore, the cross-sectional FESEM measurement reveals that the thickness of the GaN films with the growth of 60 min on the nitrided α-Al_2O_3 substrates is 300 nm, as shown in Fig. 3c, which makes us to calculate the growth rate of GaN films grown on the nitrided α-Al_2O_3 substrates at 750 ºC is ~300 nm/h.

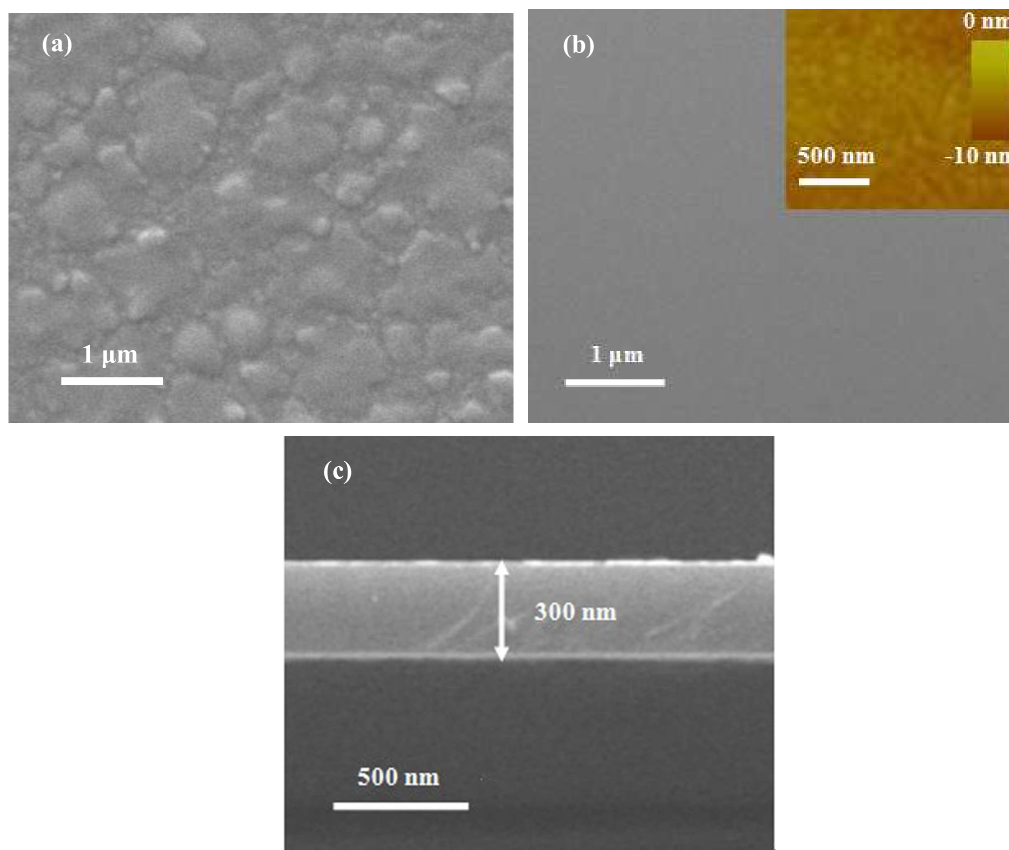

Fig. 3. FESEM images for ~300 nm-thick GaN films grown on the (a) non-nitrided, and (b) nitrided α-Al_2O_3 substrates at 750 ºC. The inserted image in Fig, 3b is the AFM image of ~300 nm-thick GaN films grown on nitrided α-Al_2O_3 substrates at 750 ºC. (c) Cross-sectional FESEM image for GaN films grown on nitrided α-Al_2O_3 substrates at 750 ºC for 60 min.

The structural properties of GaN films grown on nitrided α-Al_2O_3 substrates are studied by

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HRXRD. Fig. 4a shows the typical 2θ-θ scan for GaN films grown on nitrided α-Al_2O_3 substrates. From Fig. 4a, we can clearly identify the peaks located at 2θ=34.56° and 2θ=72.70°, which are the diffraction of GaN(0002) and GaN(0004), respectively; while the peaks observed at 2θ=41.68° and 2θ=90.58° are the diffraction of α-Al_2O_3(0006) and α-Al_2O_3(00012), respectively.[30-31] These results therefore confirm that single-crystalline GaN films have been grown on nitrided α-Al_2O_3 substrates with an out-of-plane alignment of GaN[0001]//α-Al_2O_3[0001]. Fig. 4b displays the typical φ scans of GaN(11-22) and α-Al_2O_3(1-102) planes. From Fig. 4b, one can find the six-fold rotational peaks for GaN(11-22) and three-fold rotational peaks for α-Al_2O_3(1-102). The crystal structure of α-Al_2O_3 is not the exact hexagonal structure. Therefore, there are only three-fold rotational peaks for α-Al_2O_3 (1-102).[32-33] This is in striking contrast to that of hexagonal structure GaN or AlN with six-fold rotational peaks for (1-102). The φ scans for GaN(11-22) and α-Al_2O_3(1-102) planes help to obtain an in-plane alignment of GaN[11-20]//α-Al_2O_3[1-100] between GaN films and α-Al_2O_3 substrates. This alignment reveals a lattice mismatch of 15.9% and is 30° rotated with its ideal alignment of GaN[1-100]//α-Al_2O_3[1-100] with a lattice mismatch of 0.4%, as shown in Figs. 4c-e. This may be attributed to the smaller interfacial energy in the alignment of GaN[11-20]//α-Al_2O_3[1-100], which is much easier for the nucleation of films on α-Al_2O_3 substrates when compared with that of GaN[1-100]//α-Al_2O_3[1-100].[34-37]

The crystalline quality of as-grown GaN films is characterized by X-ray rocking curves (XRCs). The full-width at half-maximums (FWHMs) of GaN (0002) and GaN (10-12) are 0.2° and 0.7° for the ~300 nm-thick GaN films grown on nitrided α-Al_2O_3 substrates, respectively, as shown in Figs. 5a and b; while the FWHMs of GaN(0002) and GaN(10-12) are 1.1° and 1.3° for the ~300 nm-thick GaN films grown on non-nitrided α-Al_2O_3 substrates, respectively. It is known that the FWHM of GaN (0002) is related to screw dislocations which are generated from the different step heights of the substrate; while the FWHM of GaN (10-12) is corresponding to pure edge and mixed dislocations that are formed during the coalescence process among the disoriented individual islands.[38-39] The dislocation density in the as-grown GaN films can be approximately calculated by the following equation:[40-41]

$$D_{dis} = \frac{\beta^2}{9b^2} \qquad (1)$$

where D_{dis} is the dislocation density, β is the FWHM value of XRC peaks, and b is the length of

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the Burger vector of the corresponding dislocation. Therefore, the dislocation densities in GaN films grown on the nitrided α-Al_2O_3 substrates and the non-nitrided α-Al_2O_3 substrates are 10^9 and 10^{11} cm^{-2}, respectively. This reveals the higher crystalline quality of GaN films grown on the nitrided α-Al_2O_3 substrates. Evidently, the nitridation process on α-Al_2O_3 substrates plays a significant role in achieving high-quality GaN films. Furthermore, the crystalline quality of GaN films achieved in this work is much better than that grown by MOCVD and MBE with the same thickness.[42] This may be ascribed to the highly energetic precursors produced by PLD, which is more beneficial to the migration of precursors on the substrates and thereby leads to the higher crystalline quality of GaN films ultimately.

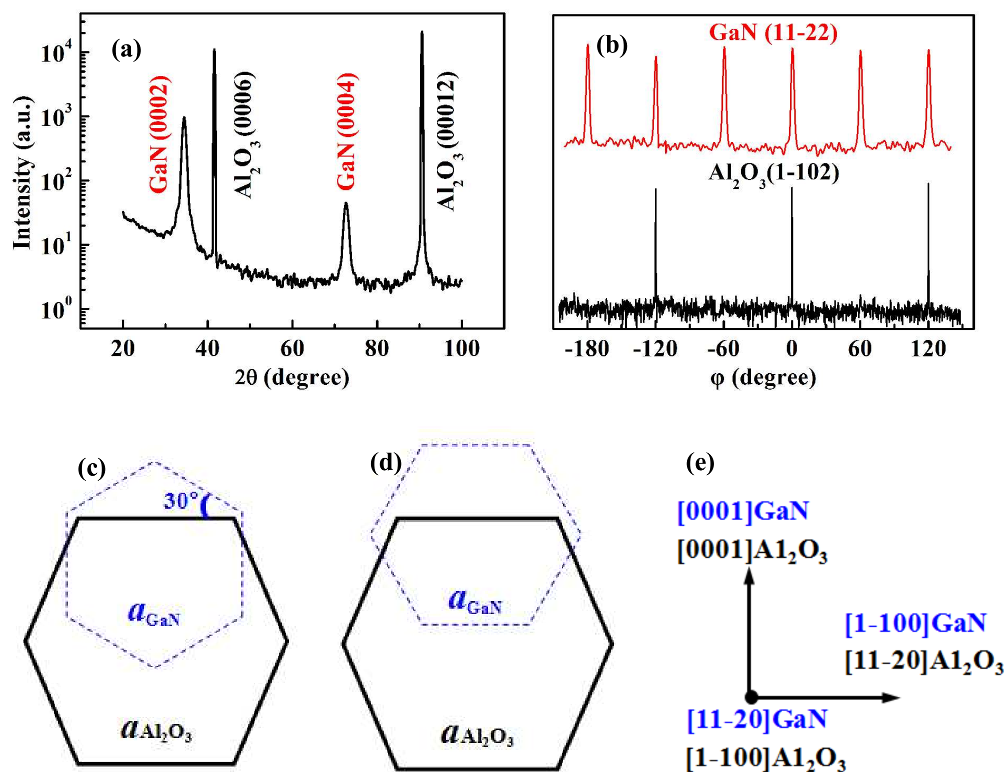

Fig. 4. (a) Typical 2θ-θ scan of ~300 nm-thick GaN films grown on nitrided α-Al_2O_3 substrates at 750 ºC, and (b) φ scans of GaN (11-22) and α-Al_2O_3 (1-102). (c) The real and (d) ideal alignment between GaN films and α-Al_2O_3 substrates. (e) The corresponding in-plane epitaxial alignments for GaN films grown on α-Al_2O_3 substrates.

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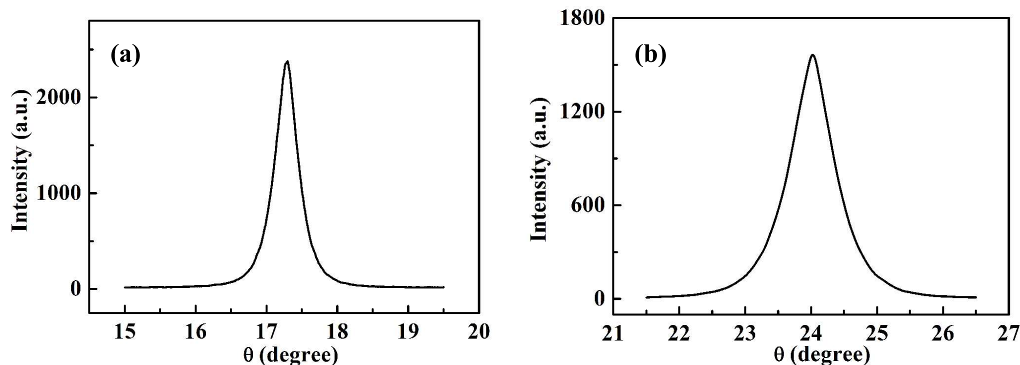

Fig. 5. Typical (a) (0002), and (b) (10-12) XRCs for ~300 nm-thick GaN films grown on nitrided α-Al_2O_3 substrates at 750 ºC.

The grazing incidence X-ray reflectivity (GIXR) is deployed to study the interfacial properties of GaN/α-Al_2O_3 hetero-interfaces. Due to the requirement of GIXR measurement,[13-14] we use the thin films for this study. Fig. 6 shows the GIXR and its simulated curves for the ~36.5 nm-thick GaN films grown on the nitrided α-Al_2O_3 substrates at 750 ºC. By fitting the curve with LEPTOS software according to the manual of Bruker D8,[15, 42-44] we find that there is a maximum of ~1.2 nm-thick interfacial layer existing between GaN films and α-Al_2O_3 substrates, and the root-mean-square (RMS) roughness of GaN films is 1.2 nm. This interfacial layer is ascribed to the AlN layer formed during the nitridation process, which is obtained from the fitted results. On the contrary, there is a interfacial layer with the thickness of 6.0 nm existing between GaN films and non-nitrided α-Al_2O_3 substrates. By fitting the results, we find that the interfacial layer may be AlGaN layer. Moreover, the RMS roughness for GaN films is 2.9 nm in this case. The formation of this interfacial layer may be ascribed to the interfacial reactions between GaN and α-Al_2O_3 substrates. We attribute these different results to the nitridation of α-Al_2O_3 substrates, which leads to the formation of a thin AlN layer.[45-47] On the one hand, this AlN layer is good for the nucleation of GaN films and enhances the migration of GaN precursors on surfaces. On the other hand, this AlN layer hampers the generation of the interfacial layer between GaN and α-Al_2O_3 substrates. On the contrary, as for GaN films grown non-nitrided α-Al_2O_3 substrates, the diffused Al atoms may react with the Ga and N plasmas. The reactions leads to the formation of the disordered interfacial layer, where dislocations are formed.[16, 42] In this case, it is difficult to grow high-quality GaN films on this interfacial layer.

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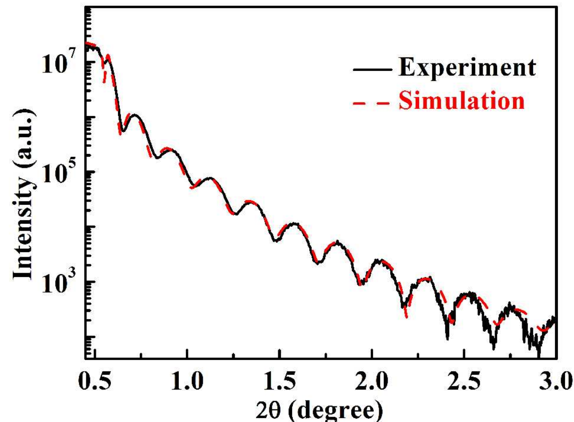

Fig. 6. GIXR curve for ~36.5 nm-thick GaN films grown on nitrided α-Al_2O_3 substrates at 750 ºC.

Cross-sectional TEM is employed to further study the interfacial properties of GaN films grown on α-Al_2O_3 substrates at 750 ºC. If the nitridation process is performed, one can clearly identify that there is only a maximum of ~1.2 nm-thick interfacial layer existing between GaN films and nitrided α-Al_2O_3 substrates, as shown in Fig. 7a. However, there is a ~6 nm-thick interfacial layer existing between GaN films and non-nitrided α-Al_2O_3 substrates, as shown in Fig. 7b. This result is a striking contrast to that with nitridation process and is well consistent with the GIXR measurement. Evidently, the nitridation process plays an important role in achieving high interfacial properties GaN/α-Al_2O_3 heterointerfaces. After carefully studying the TEM images, we obtained an in-plane alignment of GaN[1-100]//α-Al_2O_3[11-20] between GaN films and α-Al_2O_3 substrates. Meanwhile, another in-plane epitaxial relationship of GaN[11-20]//α-Al_2O_3[1-100] can be obtained from studying the direction of the electron diffraction, which agrees well with the results of RHEED measurements and XRD φ scans.[15, 48-50]

Based on these characterizations, the growth mechanism for GaN films grown on the nitrided and non-nitrided α-Al_2O_3 substrates can be deduced. The growth processes for GaN films grown on nitrided α-Al_2O_3 substrates are illustrated in Fig. 8a. The N plasmas produced by RF plasma generator would react with Al atoms on the surface and lead to the formation of the very thin AlN layer during the nitridation process. This formed AlN layer can be found in Fig. 7a, as illustrated by two white dashed lines, is good for the nucleation of GaN films and enhances the migration of GaN precursors on surface. Therefore, GaN films with smooth surface and high structural

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properties are achieved on nitrided α-Al_2O_3 substrates. Meanwhile the growth processes for GaN films grown on non-nitrided α-Al_2O_3 substrates are shown in Fig. 8b. During the initial growth, the diffused Al atoms at the substrate surface may react with the Ga plasmas and N plasmas, which results in the formation of disordered AlGaN layer. There are many dislocations formed in this layer, as illustrated by the red frames, which may propagate into the subsequent growth films, Fig. 7b. After the interfacial layer growth, the GaN films are grown by the reaction between Ga plasmas and N plasmas, as shown in Figs. 8a and b. Even though the GaN films then can be grown on this interfacial layer, the surface morphologies and structural properties for the as-grown GaN films are much poorer. Conclusively, the nitridation of α-Al_2O_3 substrates can effectively improve the surface morphologies and structural properties of as-grown GaN films by PLD.

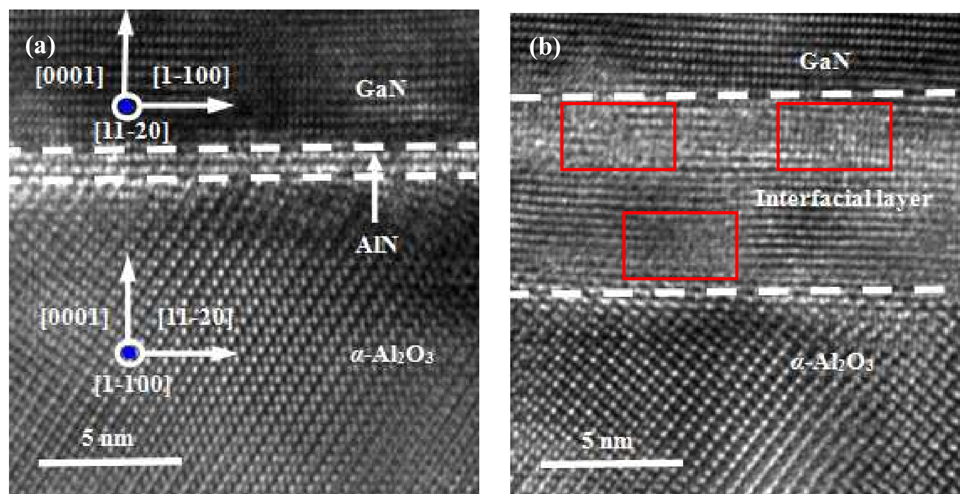
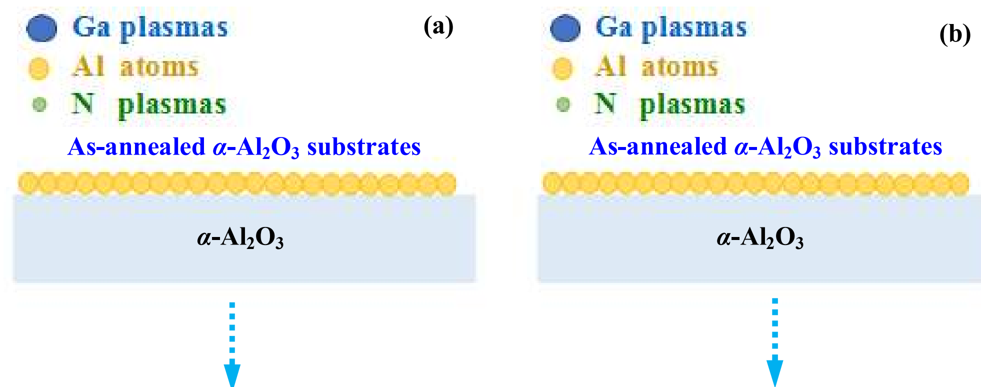


Fig. 7. Cross-sectional TEM images for GaN films grown on the (a) nitrided, and (b) non-nitrided α-Al_2O_3 substrates at 750 ºC.



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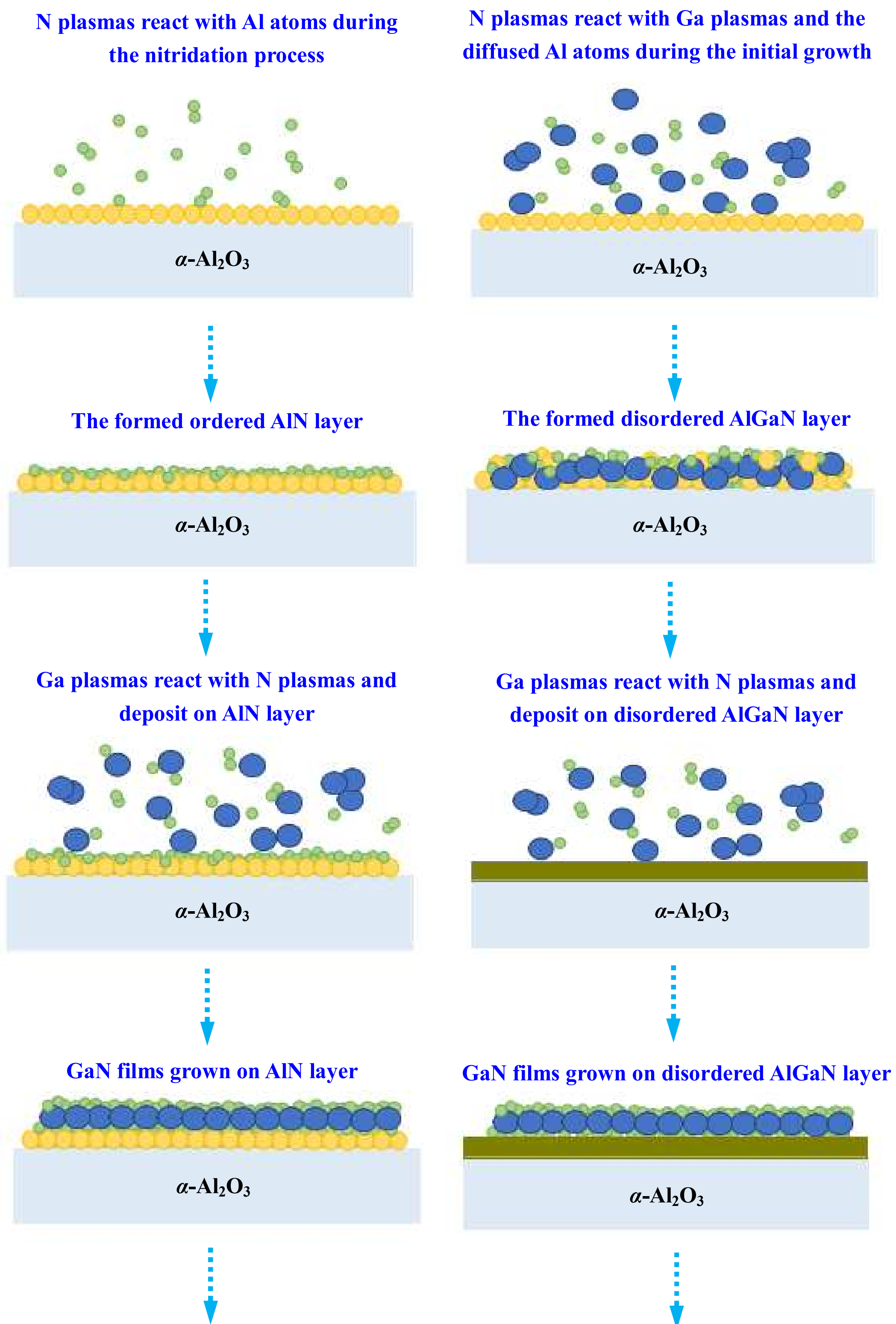

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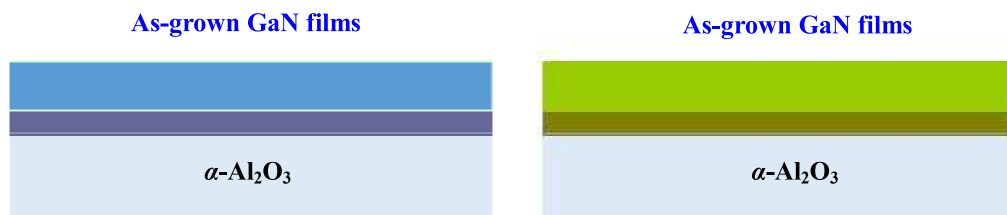

Fig. 8. Schematic diagrams for the GaN films grown on (a) nitrided, and (b) non-nitrided α-Al_2O_3 substrates, respectively.

4. Conclusions

In this work, we have systematically investigated the effect of α-Al_2O_3 substrates nitridation on the properties of GaN films grown by PLD, and proposed the growth mechanism of GaN films on nitrided α-Al_2O_3 substrates by PLD. If the nitridation process is performed on the α-Al_2O_3 substrates, the surface morphologies and structural properties of as-grown GaN films are improved significantly. We tentatively attribute this achievement of high-quality GaN films to the nitridation of the α-Al_2O_3 substrates, which leads to the formation of a thin AlN layer. On the one hand, this AlN layer is good for the nucleation of GaN films and enhances the migration of GaN precursors on surface. On the other hand, this AlN layer hampers the generation of the interfacial layer. This work brings up a broad prospect for the preparation of high-quality GaN-based devices on nitrided α-Al_2O_3 substrates by PLD.

Acknowledgements

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References

1. L. C. Wang, Y. Y. Zhang, X. Li, E. Q. Guo, Z. Q. Liu, X. Y. Yi, H. W. Zhu and G. H. Wang, *RSC Adv.*, 2013, **3**, 10934-10943.

2. R. H. Jiang, C. F. Lin, Y. C. Huang, F. H. Fan, K. C. W. J. H. Wang, P. F. Cheng and C. C. Yang, *RSC Adv.*, 2013, **3**, 13446-13450.

3. B. D. Liu, Z. E. Wang, F. Yuan, D. Benjamin, T. Sekiguchi and X. Jiang, *RSC Adv.*, 2013, **3**, 22914-22917.

4. R. Ramesh, R. Loganathan, S. S. Menon, K. Baskar and S. Singh, *RSC Adv.,* 2014, **4**, 7112-7119.

5. Y. B. Dai, Y. L. Shao, Y. Z. Wu, X. P. Hao, P. Zhang, X. Z. Cao, L. Zhang, Y. Tian and H. D. Zhang, *RSC Adv.*, 2014, **4**, 21504-21509

6. L. Zhang, Y. L. Shao, X. P. Hao, Y. Z. Wu, H. D. Zhang, S. Qu, X. F. Chen and X. G. Xu, *CrystEngComm*, 2011, **13**, 5001-5004.

7. H. D. Zhang, Y. L. Shao, L. Zhang, X. P. Hao, Y. Z. Wu, X. Y. Liu, Y. B. Dai and Y. Tian, *CrystEngComm*, 2012,**14**, 4777-4780.

8. Y. L. Shao, Y. B. Dai, X. P. Hao, Y. Z. Wu, L. Zhang, H. D. Zhang and Y. Tian, *CrystEngComm*, 2013, **15**, 7965-7969.

9. Q. L. Zhang, F. Y. Meng, P. A. Crozier, N. Newman and S. Mahajan, *Acta Materialia*, 2011, **59**, 3759-3769.

10. S. Z. Zhou, H. Y. Wang, Z. T. Lin, X. S. Hong and G. Q. Li, *Jpn. J. Appl. Phys.,* 2014, **53**, 025503.

11. T. F. Chen, Y. Q. Wang, P. Xiang, R. H. Luo, M. G. Liu, W. M. Yang, Y. Ren, Z. Y. He, Y. B. Yang, W. J. Chen, X. R. Zhang, Z. S. Wu, Y. Liu and B. J. Zhang, *Appl. Phys. Lett.*, 2012, **100**, 241112.

12. Y. J. Sun, T. J. Yu, C. Y. Jia, Z. Z. Chen, P. F. Tian, X. N. Kang, G. J. Lian and G. Y. Zhang, *Chin. Phys. Lett.*, 2010, **27**(12), 127303.

13. W. L. Wang, W. J. Yang, Z. L. Liu, Y. H. Lin, S. Z. Zhou, H. R. Qian, F. L. Gao and G. Q. Li, *CrystEngComm*, 2014, **16**, 4100-4107.

14. W. L. Wang, W. J. Yang, Z. L. Liu, Y. H. Lin, S. Z. Zhou, H. R. Qian, F. L. Gao, H. Yang and G. Q. Li, *Appl. Surf. Sci.*, 2014, **294**, 1-8.

RSC Advances Accepted Manuscript

15. H. Yang, W. L. Wang, Z. L. Liu and G. Q. Li, *CrystEngComm*, 2013, **15**, 7171-7176.

16. H. Yang, W. L. Wang, Z. L. Liu, W. J. Yang and G. Q. Li, *CrystEngComm*, 2014, **16**, 3148-3154.

17. W. L. Wang, Z. L. Liu, W. J. Yang, Y. H. Lin, S. Z. Zhou, H. R. Qian, H. Y. Wang, Z. T. Lin and G. Q. Li, *Mater. Lett.*, 2014, **129**, 39-42.

18. H. Yang, W. L. Wang, Z. L. Liu and G. Q. Li, *J. Phys. D: Appl. Phys.*, 2013, **46**, 105101.

19. J. A Greer, J. Phys. D: Appl. Phys., 2014, **47**, 034005.

20. K.W. Mah, E. McGlynn, J. Castro, J. G. Lunney, J. P. Mosnier, D. O'Mahony and M. O. Henry, *J. Cryst. Growth*, 2001, **222**, 497-502.

21. T. F. Huang, A. Marshall, S. Spruytte and J. S. Harris Jr. *J. Cryst. Growth*, 1999, **200**, 362-367.

22. D. Feiler, R. S. Williams, A. A. Talin, H. Yoon and M. S. Goorsky, *J. Cryst. Growth*, 1997, **171**, 12-20.

23. W. L. Wang, W. J. Yang, Z. L. Liu, Y. H. Lin, S. Z. Zhou, H. R. Qian, F. L. Gao, L. Wen, S. G. Zhang and G. Q. Li, *J. Mater. Sci.*, 2014, **49** (9), 3511-3518.

24. R. D. Vispute, V. Talyansky, R. P. Sharma, S. Choopun, M. Downes, T. Venkatesan, K. A. Jones, A. A. Iliadis, M. Asif Khan and J. W. Yang, *Appl. Phys. Lett.,* 1997, **71**, 102-104.

25. P. Sanguino, M. Niehus, L.V. Melo, R. Schwarz, S. Koynov, T. Monteiro, J. Soares, H. Alves and B.K. Meyer, *Solid-State Electron.*, 2003, **47**, 559-563.

26. C. Vinegonia, M. Cazzanelli, A. Trivelli, G. Mariotto, J. Castro, J. G. Lunney and J. Levy, *Surf. Coat. Tech.*, 2000, **124**, 272-277.

27. A. P. McKiernan and J. P. Mosnier, *Appl. Surf. Sci.*, 2002, **197-198**, 325-330.

28. C. Gaire, F. Tang and G. C. Wang, *Thin Solid Films*, 2009, **517**, 4509-4514.

29. A. Ichimiya and P. I. Cohen, *Reflection high-energy electron diffraction*, Cambridge University press, 2004.

30. D. Cole , J. G. Lunney, F. P. Logue, J. F. Donegan and J. M. D. Coey, *Mater. Sci. Eng. B*, 1997, **48**, 239-243.

31. M. Kumar, T. N. Bhat, M. K. Rajpalke, B. Roul, A.T. Kalghatgi and S. B. Krupanidhi, *J. Alloy. Compd.*, 2012, **513**, 6-9.

32. Z. J. Xu, *Semiconductor testing and analysis*, 2 nd ed., Beijing: Science press, 2007.

RSC Advances Accepted Manuscript

33. X. P. Zhang, *Study of Sapphire Crystal Growth and Its Application on LED*, Hunan University, 2009.

34. M. Sumiya, T. Chikyow, T. Sasahara, K. Yoshimura, J. Ohta, H. Fujioka, S. Tagaya, H. Ikeya, H. Koinuma and S. Fuke, *Jpn. Appl. Phys. Part 1*, 2002, **41**, 5038-5041.

35. J. Ohta, H. Fujioka, M. Sumiya, H. Koinuma and M. Oshima, *J. Cryst. Growth*, 2001, **225** 73-78 (2001).

36. S. Inoue, K. Okamoto, N. Matsuki, T. W. Kim and H. Fujioka, *J. Cryst. Growth*, 2006, **289** 574-577.

37. U. W. Pohl, *Epitaxy of Semiconductors*, 1st ed. New York: Springer, 2013.

38. S. T. Li, F. Y. Jiang, G. H. Fan, W. Q. Fang and L. Wang, *Phys. B*, 2007, **391**, 169-173.

39. Y. J. Liu, T. Y. Tsai, C. H.Yen, L. Y. Chen, T. H. Tsai, C. C. Huang, T. Y. Chen, C. H. Hsu and W. C. Liu, *Opt. Express*, 2010, **18**(3), 2729-2742.

40. W. L. Wang, H. Yang and G. Q. Li, *J. Mater. Chem. C*, 2013, **1**, 4070-4077.

41. M. A. Moram and M. E. Vickers, *Rep. Prog. Phys.*, 2009, **72**, 036502.

42. W. L. Wang, H. Yang and G. Q. Li, *CrystEngComm*, 2013, **15**, 2669-2674.

43. L. G. Parratt, *Phys. Rev.*, 1994, **95**, 359-369.

44. L. Névot and P. Croce, *Rev. Phys. Appl.,* 1980, **15**, 761-779.

45. F. L. Gao, Y. F. Guan, J. L. Li, J. N. Gao, J. Q. Guo and G. Q. Li, *J. Mater. Res.,* 2013, **28** (9), 1239-1244.

46. M. Sawadaishi, S. Taguchi, K. Sasaya and T. Honda, *J. Cryst. Growth*, 2009, **311**, 1994-1996.

47. T. Honda, H. Yamamoto, M. Sawadaishi, S. Taguchi and K. Sasaya, *J. Cryst. Growth*, 2009, **311**, 2844-2846.

48. Y. H. Ra, R. Navamathavan, J. H. Park and C. R. Lee, *CrystEngComm*, 2013, **15**, 1874-1881.

49. Y. H. Ra, R. Navamathavan and C. R. Lee, *CrystEngComm*, 2012, **14**, 8208-8214.

50. W. L. Wang, W. J. Yang, Z. L. Liu, Y. H. Lin, S. Z. Zhou, H. R. Qian, H. Y. Wang, Z. T. Lin and G. Q. Li, *RSC Adv.*, 2014, **4**, 27399-27403.

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