Phys. Status Solidi C, **9**, No. 3–4, 519–522 (2012) / **DOI** 10.1002/pssc.201100499

physica status solidi c
www.pss-c.com
current topics in solid state physics

Growth of GaN and AlGaN on (100) β-Ga_2O_3 substrates

Shun Ito*[, 1], Kenichiro Takeda[1], Kengo Nagata[1], Hiroki Aoshima[1], Kosuke Takehara[1], Motoaki Iwaya[1], Tetsuya Takeuchi[1], Satoshi Kamiyama[1], Isamu Akasaki[1,3], and Hiroshi Amano[2,3]

[1] Faculty of Science and Technology, Meijo University, Nagoya 468-8502, Japan
[2] Department of Electrical Engineering and Computer Science, Nagoya University, Nagoya 464-8603, Japan
[3] Akasaki Research Center, Nagoya University, Nagoya 464-8603, Japan

Received 1 August 2011, revised 8 November 2011, accepted 8 November 2011
Published online 25 January 2012

Keywords β-Ga_2O_3(100), thermal annealing, GaN, AlGaN, MOVPE

* Corresponding author: e-mail 103434008@ccalumni.meijo-u.ac.jp

The crystalline quality of GaN and $Al_{0.08}Ga_{0.92}N$ epitaxial layers on (100) β-Ga_2O_3 substrates was significantly improved by the facet-controlled growth method. The facets were controlled by changing the nitrogen ambient thermal annealing temperature. We demonstrated the high-crystalline-quality GaN and $Al_{0.08}Ga_{0.92}N$ on β-Ga_2O_3 substrates, which were comparable to GaN and AlGaN on sapphire substrates using low-temperature buffer layers. This method is useful for the fabrication of vertical-type ultraviolet (UV) light-emitting diodes (LEDs) on β-Ga_2O_3 substrates.

© 2012 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

1 Introduction Group III-nitride-based UV LEDs are expected in many applications such as excitation sources of white LED lamps, material processing, healthcare field, and sterilization. There have been many reports on high-efficiency UV LEDs on sapphire substrates [1, 2]. However, the wall plug efficiency of UV LEDs is still lower than that of GaInN-based blue LEDs [3]. One of the major causes of such low-wall-plug efficiency in the UV LEDs is the current crowding effect caused by the combination of insulating sapphire substrates and an n-type AlGaN cladding layer with a relatively higher sheet resistance than that of GaN. One of the best methods to solve this problem is to use a vertical conductive device structure. Thus far, GaInN-based vertical visible and near-UV LEDs were grown on GaN and SiC conductive substrates [4, 5]. However, there is no report on AlGaN-based vertical UV LEDs on conductive transparent substrates. β-Ga_2O_3 is one of the most attractive substrates for AlGaN-based vertical UV LEDs. It has a transparency of up to 260 nm and n-type high conductivity [6]. These properties can lead to a small absorption of UV light in β-Ga_2O_3 and make it possible to realize vertical conductive LEDs. Therefore, β-Ga_2O_3 is a promising material for the substrate of vertical UV LEDs. Several groups have reported the GaN epitaxial growth on β-Ga_2O_3 and GaInN-based blue LEDs [7, 8]. Moreover, our group has reported AlGaN with mirror surfaces on β-Ga_2O_3 using low-temperature GaN buffer layers (LT-GaN), which is essential for UV LEDs [9]. However, the crystalline quality of these epitaxial layers was poorer than that on sapphire substrates. Thus, high-quality AlGaN films on β-Ga_2O_3 substrates have been urgently required to achieve high-performance UV LEDs. In this study, to obtain high-crystalline-quality GaN and AlGaN epitaxial layers on (100) β-Ga_2O_3 substrates, GaN and AlGaN were grown using facet layers reported previously [10]. The thermal annealing temperature of β-Ga_2O_3 substrates was used as a parameter to control the facet formation.

2 Experimental procedure In this study, crystal growth was performed by metal organic vapor phase epitaxy. The aluminium and gallium sources were trimethylaluminium and trimethylgallium, respectively. The nitrogen source was NH_3. Moreover, the thermal annealing and growth of GaN and $Al_{0.08}Ga_{0.92}N$ were carried out in nitrogen ambient, because β-Ga_2O_3 substrates were etched using hydrogen. The GaN and $Al_{0.08}Ga_{0.92}N$ growths were carried out using LT-GaN. $Al_xGa_{1-x}N$ (0001) films grow epitaxially on (100) β-Ga_2O_3 substrates with an in-plane epitaxial relationship of $Al_xGa_{1-x}N$ [1-100]‖β-Ga_2O_3[001] [11].

WILEY ONLINE LIBRARY

© 2012 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

Figure 1 shows the timing charts of the growth temperatures of GaN and $Al_{0.08}Ga_{0.92}N$ without facet $Al_xGa_{1-x}N$ layers. After performing the thermal annealing for 3 min at 1100 °C and deposition of LT-GaN at 550 °C, GaN and $Al_{0.08}Ga_{0.92}N$ of approximately 2.0-μm-thick were grown at 1080 °C.

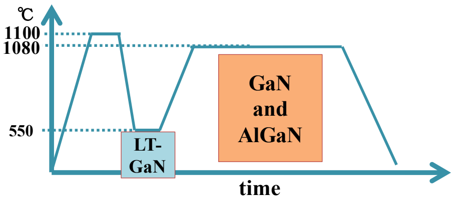

Figure 1 Timing chart of the growth temperatures of GaN and AlGaN.

Figure 2 shows the timing charts of the growth temperatures of GaN and $Al_{0.08}Ga_{0.92}N$ grown with facet-$Al_xGa_{1-x}N$ layers. The thermal annealing for 3 min and deposition of LT-GaN at 550 °C were performed in the same manner as for the samples without facet-$Al_xGa_{1-x}N$ layers. In addition, approximately 300-nm-thick facet-GaN or $Al_{0.08}Ga_{0.92}N$ was grown at 1020 and 950 °C. The growth temperatures of the GaN and $Al_{0.08}Ga_{0.92}N$ facets were lower than those of conventional GaN and $Al_{0.08}Ga_{0.92}N$ layers to enhance island growth. Finally, GaN and $Al_{0.08}Ga_{0.92}N$ of approximately 2.0-μm-thick were grown on facet-GaN and $Al_{0.08}Ga_{0.92}N$ at 1080 °C. In this case, the thermal annealing temperature was changed from 600 to 1100 °C.

Samples were characterized by atomic force microscopy (AFM), scanning electron microscopy (SEM), X-ray rocking curve (XRC), and photoluminescence (PL) measurement using a He-Cd laser (325 nm) at room temperature.

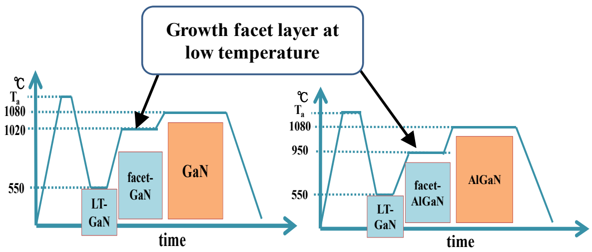

Figure 2 Timing charts of growth temperatures of (a) GaN and (b) AlGaN using facet layers.

3 Results

3.1 Impact of thermal annealing temperature on β-Ga_2O_3 Figure 3 shows AFM images of (a) (100) β-Ga_2O_3 substrate and (100) β-Ga_2O_3 substrates annealed at (b) 600 °C, (c) 800 °C, (d) 950 °C, and (e) 1100 °C for 3 min. From these figures, the surface roughness of β-Ga_2O_3 increases with increasing annealing temperature. By annealing at 1100 °C, the root mean square (RMS) roughness was increased from 0.20 to 14 nm.

3.2 Fabrication of facet-GaN Figure 4 shows plan-view SEM images of samples after substrate annealing and subsequent deposition of LT-GaN at 550 °C and growth of approximately 300-nm-thick GaN facet layers at 1020 °C. From these figures, the GaN islands with inclined facets were grown, and the area of the surface covered by the facets was controlled by adjusting the thermal annealing temperature.

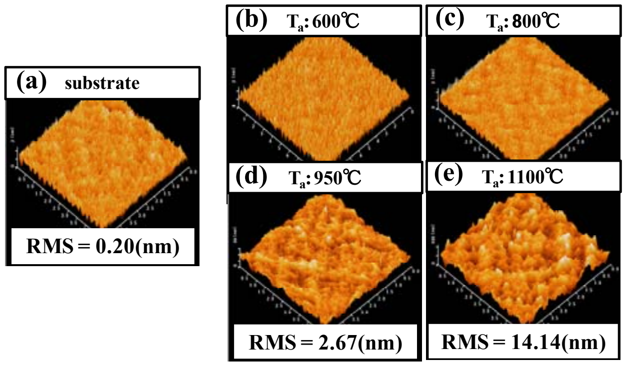

Figure 3 Plan-view AFM images of (a) (100) β-Ga_2O_3 substrate and (100) β-Ga_2O_3 substrate thermal-annealed at (b) 600 °C, (c) 800 °C, (d) 950 °C, and (e) 1100 °C for 3 min.

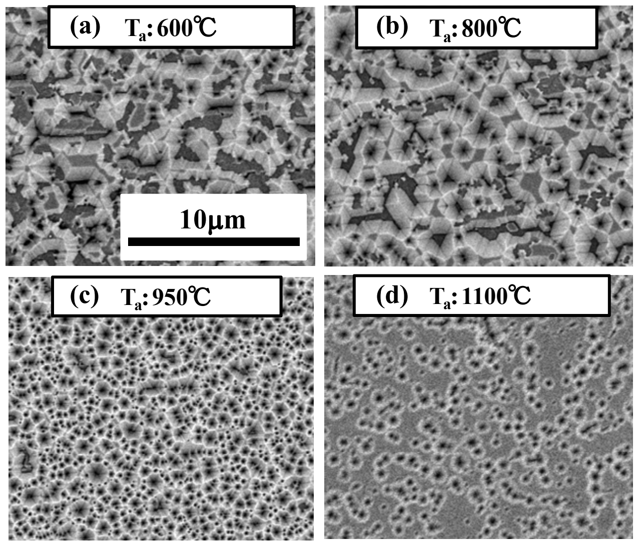

Figure 4 Plan-view SEM images of substrates thermal-annealed at (a) 600 °C, (b) 800 °C, (c) 950 °C, and (d) 1100 °C followed by deposition of low-temperature GaN buffer layer and growth of approximately 300-nm-thick GaN layer at 1020 °C.

3.3 Growth of GaN on facet layers Figure 5 shows plan-view SEM images of approximately 2.0-μm-thick GaN layers grown at 1080 °C on each facet-GaN sample. The surfaces of the GaN films on thermal-annealed substrates at (a) 600 °C and (b) 800 °C were not smooth. However the surfaces of the GaN films on samples thermal-annealed at (c) 950 °C and (d) 1100 °C were smooth and crack-free.

© 2012 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

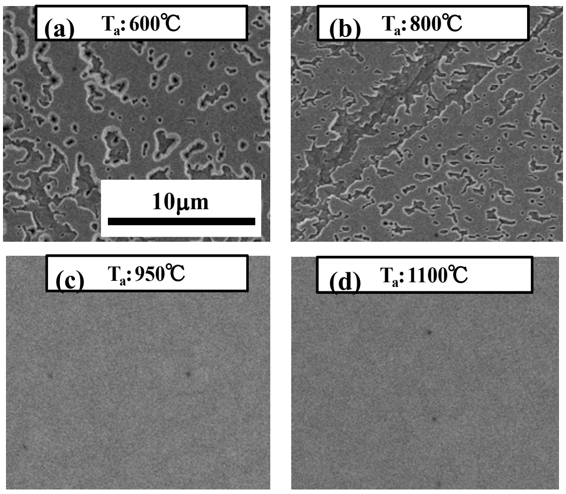

Figure 5 Plan-view SEM images of 2.0-μm-thick GaN grown at 1080 °C on substrates thermal-annealed at (a) 600 °C, (b) 800 °C, (c) 950 °C, and (d) 1100 °C followed by growth of approximately 300-nm-thick GaN facet layers at 1020 °C.

Figure 6 shows the full width at half maximum (FWHM) of XRC of GaN. Figure 4 indicates that the facet-GaN covered the whole surface when the thermal annealing temperature was 950 °C, and the flat surfaces remain when it was higher and lower than 950 °C. As mentioned in reference [10], the dislocations existing in the facet regions bend and do not glide to the surface. Therefore, the crystalline quality of GaN was highest when the annealing temperature is 950 °C. The formation of facets was related to the adequate roughness of the β-Ga_2O_3 substrates. Further evaluation is necessary.

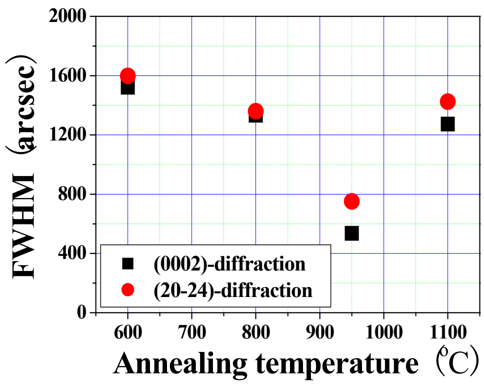

Figure 6 XRC FWHM of GaN for various thermal annealing temperatures.

Figure 7 shows the comparison of XRC FWHM between the GaN without and with the facet-GaN layer by thermal annealing at 950 °C. The threading dislocation densities of the GaN films were estimated from the FWHM values [12]. The calculated dislocation densities of GaN without and with the facet-GaN layer were 1.9x10^{10} and 2.5x10^{9} cm^{-2}, respectively. The threading dislocation density of GaN on (100) β-Ga_2O_3 substrate was successfully decreased by almost one order of magnitude. Figure 8 shows the PL spectra of the GaN with and without the facet-GaN layer. These results indicated that the facet-GaN layer improves the crystalline quality and optical property of GaN on (100) β-Ga_2O_3 substrate.

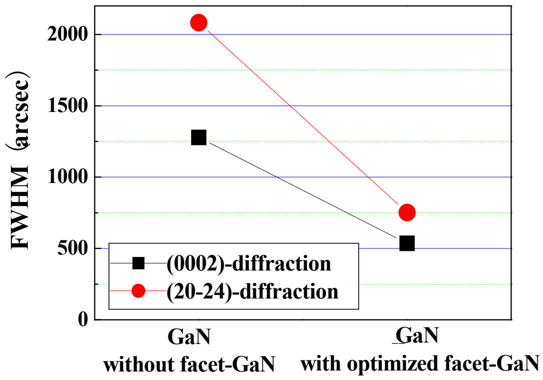

Figure 7 XRC FWHM of GaN without and with facet sample formed by thermal annealing at 950 °C.

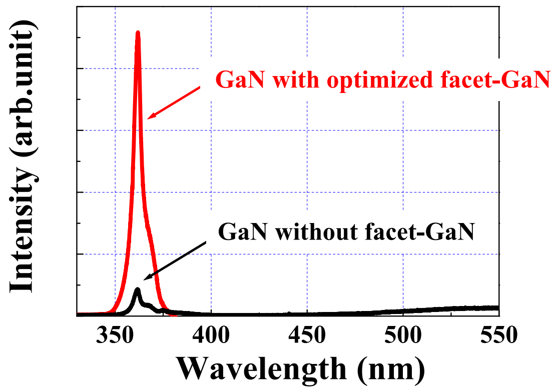

Figure 8 PL spectra of GaN without and with facet sample formed by thermal annealing at 950 °C.

3.4 Result of $Al_{0.08}Ga_{0.92}N$ The $Al_{0.08}Ga_{0.92}N$ was grown under the same growth conditions of GaN. Only the facet-$Al_{0.08}Ga_{0.92}N$ growth temperature was changed to 950 °C. Figure 9 shows a plan-view SEM images of facet-$Al_{0.08}Ga_{0.92}N$ and an approximately 2.0-μm-thick $Al_{0.08}Ga_{0.92}N$ layer grown on facet-$Al_{0.08}Ga_{0.92}N$ samples at 1080 °C. Moreover, facet-$Al_{0.08}Ga_{0.92}N$ made it possible to obtain a smooth surface.

Figure 10 shows the XRC FWHM of $Al_{0.08}Ga_{0.92}N$ without and with the facet-$Al_{0.08}Ga_{0.92}N$ layer. The calculated dislocation densities of GaN without and with the facet-GaN layer were 2.6x10^{10} and 4.9x10^{9} cm^{-2}, respectively.

 © 2012 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

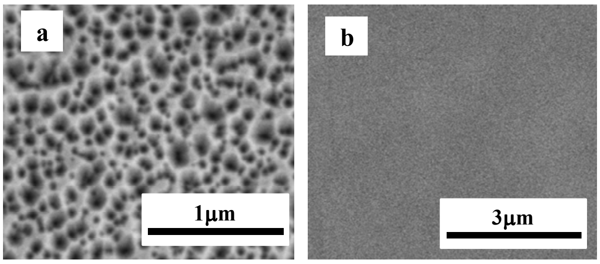

Figure 9 Plan-view SEM images of (a) facet-$Al_{0.08}Ga_{0.92}N$ and (b) approximately 2.0-μm-thick $Al_{0.08}Ga_{0.92}N$ layer grown on facet-$Al_{0.08}Ga_{0.92}N$ samples at 1080 °C.

Figure 11 shows the PL spectra of the samples without and with the facet-$Al_{0.08}Ga_{0.92}N$ layer. These results show that the facet-$Al_{0.08}Ga_{0.92}N$ layer is useful for growing high-quality $Al_{0.08}Ga_{0.92}N$ on (100) β-Ga_2O_3 substrate.

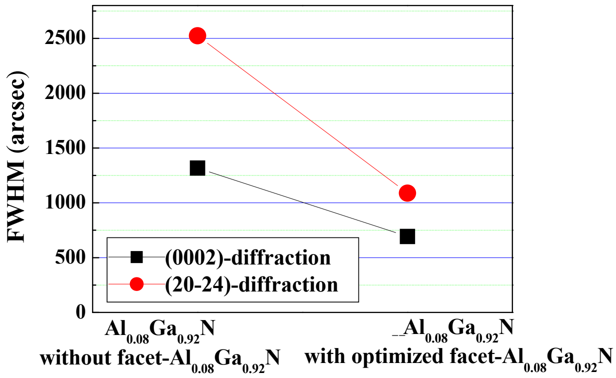

Figure 10 XRC FWHM of $Al_{0.08}Ga_{0.92}N$ without and with facet-$Al_{0.08}Ga_{0.92}N$ formed by substrate thermal annealing at 950 °C.

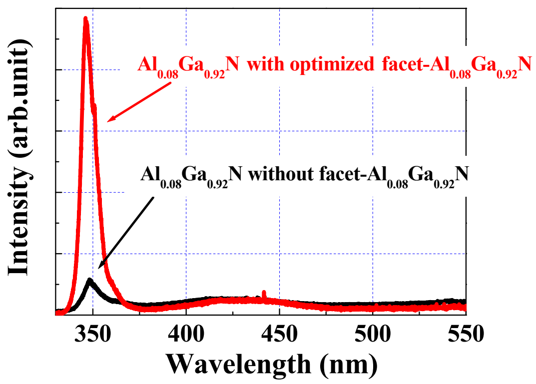

Figure 11 PL spectra of $Al_{0.08}Ga_{0.92}N$ without and with facet sample formed by thermal annealing at 950 °C.

4 Conclusion The thermal annealing of β-Ga_2O_3 substrate at 950 °C led to the form ation of GaN and $Al_{0.08}Ga_{0.92}N$ having an inclined facet covering whole surfaces, resulting in high-quality GaN and $Al_{0.08}Ga_{0.92}N$ on β-Ga_2O_3 substrates. The facet-controlled method with β-Ga_2O_3 substrates is useful for high-efficiency vertical UV LEDs in the future.

Acknowledgements This work was partially supported by a Grant-in-Aid for Scientific Research in Priority Area "Optoelectronics Frontier by Nitride Semiconductor-Ultimate Utilization of Nitride Semiconductor Material Potential" (No. 18069011) of the Ministry of Education, Culture, Sports, Science and Technology.

References

[1] H. Tsuzuki, F. Mori, K. Takeda, T. Ichikawa, M. Iwaya, S. Kamiyama, H. Amano, I. Akasaki, H. Yoshida, M. Kuwabara,Y. Yamashita, and H. Kan, Phys. Status Solidi A **206**, 1199 (2009).

[2] C. Pernot, M. Kim, S. Fukahori, T. Inazu, T. Fujita, Y. Nagasawa, A. Hirano, M. Ippommatsu, M. Iwaya, S. Kamiyama, I. Akasaki, and H. Amano, Appl. Phys. Express **3**, 061004 (2010).

[3] X. A. Cao and S. D. Arthur, Appl. Phys. Lett. **85**, 3971 (2004).

[4] J. Edmond, A. Abare, M. Bergman, J. Bharathan, K. L. Bunker, D. Emerson, K. Haberern, J. Ibbetson, M. Leung, P. Russel, and D. Slater, J. Cryst. Growth **272,** 242 (2004).

[5] Y. Narukawa, M. Ichikawa, D. Sanga, M. Sano, and T. Mukai, J. Phys. D: Appl. Phys. **43,** 354002 (2010).

[6] H. Aida, K. Nishiguchi, H. Takeda, N. Aota, K. Sunakawa, and Y. Yaguchi, Jpn. J. Appl. Phys. **47**, L11 (2008).

[7] K. Shimamura, E. G. Villora, K. Domen, K. Yui, K. Aoki, and N. Ichinose, Jpn. J. Appl. Phys. **44**, L7 (2005).

[8] X. Zi-Li, Z. Rong, X. Chang-Tai, X. Xiang-Qian, H. Ping, L. Bin, Z. Hong, J. Ruo-Lian, S. Yi, and Z. You-Dou, Chin. Phys. Lett. **25**, 2185 (2008).

[9] Y. Kawai, S. Ito, K. Takeda M. Iwaya, S. Kamiyama, H. Amano, and I. Akasaki, Ext. Abst. of ISPlasma 2010, **PB-56B** (2010).

[10] S. Nitta, J. Yamamoto, Y. Koyama, Y. Ban, K. Wakao, and K. Takahashi, J. Cryst. Growth **272**, 438 (2004).

[11] S. Ohira, N. Suzuki, H. Minami, K. Takahashi, T. Araki, and Y. Nanishi, Phys. Status Solidi C **4**, 2306 (2007).

[12] S. R. Lee, A. M. West, A. A. Allerman, K. E. Waldrip, D. M. Follstaedt, P. P. Provencio, and D. D. Koleske, Appl. Phys. Lett. **86**, 241904 (2005).

© 2012 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim