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# Abruptness Improvement of the Interfaces of AlGaN/GaN Superlattices by Cancelling Asymmetric Diffusion

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Interfacial abruptness plays a critical role in affecting the quantum confinement effect in heterostructures. Here, we accurately determine the inter-diffusion depth across the AlGaN/GaN interfaces and propose a simple blocking scheme to effectively improve the superlattice abruptness. It is found that the Al diffusion depth at the upper and lower interfaces of the AlGaN barrier appears considerably asymmetric. Such difference leads to the gradient interfacial region and the asymmetric quantum well shape. A pair of ultra-thin blocking layers is introduced to the GaN/AlGaN interface to block the Al downward diffusion. After the blocking treatment, the interfacial abruptness is improved and the light emission intensity from the superlattice can be effectively enhanced. © 2013 The Japan Society of Applied Physics

## 1. Introduction

III–nitrides have been important and promising materials for the fabrication of short wavelength optical and high power electronic devices.[1–4] Due to its wide band gap and strong polarizability, AlGaN system has recently attracted broad concerns in both advanced nano-structured materials and optoelectronic devices. In general, functional heterostructures, e.g., multiple quantum wells and superlattices, have been widely used in device fabrication owing to their distinct virtues such as lower threshold current density, lower non-radiative recombination rate, and reduced sensitivity to temperature.[5,6] Particularly, the AlGaN/GaN heterostructure has been an important building block in constructing nitride-based devices for wide applications, due to its excellent properties, e.g., high conduction band offset, large longitudinal (LO) phonon energy, and ultra-fast carrier and inter-subband relaxation.[7–9] As the main component of the active layers in device structures,[10–13] the achievement of high quality AlGaN/GaN superlattices (SLs) becomes extremely crucial in achieving high performance of the AlGaN-based optoelectronic devices.

In principles, the most critical part of a SL is the interfacial region between heterolayers, which plays the dominant role in deciding the quality, function and property of entire SLs. In order to achieve abrupt interface, some have introduced the interruption treatment with ammonia flow between the growth of well and barrier to improve the InGaN/GaN heterostructures.[14] Although the growth interruption technique has also been applied to the AlGaN/GaN structures,[15] the diffusion of Al element across the interface at high growth temperature intrinsically exists and is difficult to cancel. This diffusion governs the final quality of the interface and act as the origin causing the asymmetry of different types of interfaces. How to improve the interfacial abruptness against high temperature diffusion has become a challenge in the fabrication of nitride based SLs.

In this work, the inter-diffusion of Al across the AlGaN/GaN interface has been systematically studied and a concept of ultra-thin blocking layer is proposed to effectively minimize the high temperature diffusion. The group-III sources were shut off at different interfaces between the GaN well and AlGaN barrier layers and various time duration of the $NH_3$ flow in this growth interruption were carried out. With X-ray diffraction (XRD) and high resolution transmission electron microscopy (HRTEM), it was found that the interfacial abruptness is highly associated with the time of growth interruption. The sharpness of the GaN/AlGaN interface and the AlGaN/GaN interface appears distinctly asymmetric. A pair of ultra-thin blocking layers was then introduced to the GaN/AlGaN interface to block the Al downward diffusion. With these blocking layers, the interfacial abruptness can be considerably improved and the light emission intensity from the SLs can be effectively enhanced.

## 2. Experimental Methods

The AlGaN/GaN SLs structures were grown by metal organic chemical vapor deposition (MOCVD) with a Thomas Swan closely coupled showerhead (CCS) reactor, which is designed especially for reducing the undesired gas phase parasitic reaction. Trimethylgallium (TMGa), trimethylaluminium (TMAl), and ammonia ($NH_3$) were used as the precursors for Ga, Al, and N, respectively. $H_2$ and $N_2$ were used as carrier and bubble gases. Prior to the GaN/AlGaN SLs growth, thermal cleaning of the (0001)-oriented sapphire substrate was carried out under hydrogen ambient at 1050 °C for 10 min to remove native oxide from the surface. Then a 2-µm-thick underlying GaN template was grown after the deposition of a 20-nm-thick low-temperature GaN nucleation layer. Finally, the 20 periods of GaN/$Al_{0.4}Ga_{0.6}N$ SLs were grown at 1050 °C. The thicknesses of the $Al_{0.4}Ga_{0.6}N$ barrier and the GaN well were set to be about 3 and 5 nm, respectively. We used a thicker GaN well for the study of the interfacial abruptness and inter-diffusion, avoiding the interference between the upper and lower interfaces. The growth interruption was operated by closing the group-III sources and maintaining $NH_3$ flow at the AlGaN/GaN interface (transition from the AlGaN layer to the GaN layer) and/or the GaN/AlGaN interface (transition from the GaN layer to the AlGaN layer), as shown schematically in Fig. 1. The duration of the growth interruption in sample A, was set to be 10 s at both interfaces. In sample B, the interruption was used uniquely at the AlGaN/GaN for 10 s and the GaN/AlGaN was grown in the continuous growth mode (0 s interruption). For our compar-

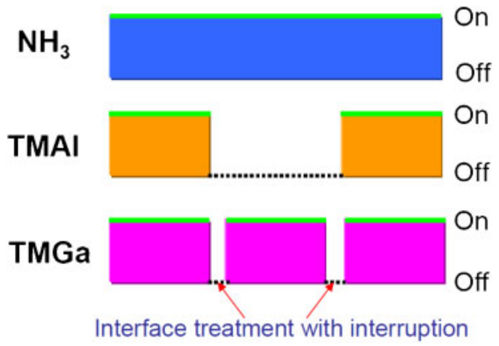

**Fig. 1.** (Color online) Schematic procedure of source materials in the growth of GaN/AlGaN SLs structure with growth interruption at different interfaces.

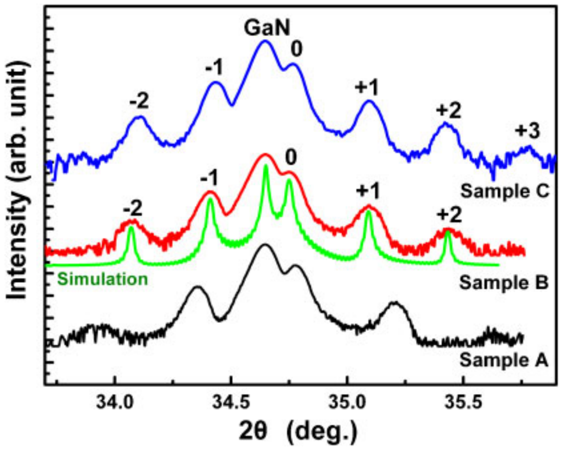

**Fig. 2.** (Color online) (0002) $\omega$–$2\theta$ scan of the GaN/AlGaN SLs for samples A, B, and C. The obvious improvement of the interfacial abruptness in sample B is achieved with the alternative interruption scheme (10 s/0 s), compared to that to sample A (10 s/10 s). Furthermore, the blocking layer employed in sample C significantly improves the interfacial abruptness, which shows the +3 order satellite.

ison, all the other growth parameters for these GaN/$Al_{0.4}Ga_{0.6}N$ SLs samples were kept the same.

In order to improve the interface abruptness, a novel scheme of ultra-thin blocking layers was employed to treat the GaN/AlGaN interface in sample C. A lower-Al-content blocking layer (B1) was deposited right above the GaN layer. Then another high-Al-content layer, called compensation layer (B2), was grown to inject Al into the B1 layer (increasing the average Al content). Thus, both B1 and B2 layers could possess the same Al content as that of the upper AlGaN barrier layer. At the topmost surface, a thin GaN cladding layer in 5 nm thick was put to protect the whole SLs structure beneath.

The structural quality and the abruptness of the GaN/AlGaN interfaces were evaluated by XRD with a Bede QC200 system. Cross-sectional high-angle annular dark-field (HAADF) scanning transmission electron microscopy (STEM) and HRTEM was performed using a TECNAI F30 system operated at 200 kV. The cross-section samples were prepared by a conventional combination of mechanical grinding, dimpling, and ion milling. The $Z$-contrast images of the SLs for elements Al, Ga, N, and O were scanned with STEM to clarify the different structural and chemical condition at the AlGaN/GaN and GaN/AlGaN interfaces. To accurately determine the diffusion depth of metal elements across the interface, the Auger electron spectroscopy (AES) was employed to profile the chemical composition of the SLs with a PHI-660 system. AES has been proved to be a powerful tool for detecting local information from the surface layers within 2 nm. The optical properties were investigated by photoluminescence (PL) measurements at room temperature. PL spectra were excited by the 248 nm line of a KrF excimer laser and the emitted photons were collected by a spectrometer (Avantes 2048 × 12).

## 3. Results and Discussion

First, various interruption conditions were used to modify both the GaN/AlGaN and AlGaN/GaN interfaces and the structural information was measured by XRD $\omega$–$2\theta$ scans of the symmetrical (0002) Bragg reflection. The results showed that the interface quality is closely associated with the duration of interruption. Interruption with only ammonia flow less than 10 s would be better for the interface smoothening. However, due to the different characteristic between these two types of interfaces, the uniform interruption just leads to the inequality in the abruptness of either interface, which will be further discussed below in detail. In this regard, we propose the first interfacial treatment scheme to deal with different types of interfaces, called alternative interruption. For example, in sample B, an interruption with 10 s was employed for the AlGaN/GaN interface whereas continuous growth (0 s) was used at the GaN/AlGaN interface. The XRD scan in Fig. 2 shows clearly that the SL satellites from −2 to +2 order become visible and distinctly sharp for sample B, while for sample A the satellites are broad and have less orders. This confirms that the interface quality of sample B is effectively improved. The reason why we skipped the growth interruption treatment for the GaN/AlGaN interface is because that the etching effect by ammonia during the interruption is significant at this interface (GaN surface),[16] which will deteriorate the interface smoothness and turn it rough. On the other hand, the interval distance between adjacent satellites is closely associated with the GaN/AlGaN thickness. One can see that the thickness of the GaN layer in sample A is less than that in sample B. This suggests that the longer interruption time may lead to the GaN layer decomposition at the GaN/AlGaN interface and hence, the thickness is reduced. Meanwhile the Al atoms at this interface will undergo a significant downward diffusion into GaN well layer. Both these two reactions at the GaN/AlGaN interface during the interruption only bring about negative influences on the interfacial sharpness. Therefore, the removal of interruption at the GaN/AlGaN interface can improve the SL quality.

After interruption treatment, could the interfaces of the AlGaN/GaN SLs become abrupt enough? To observe the interface structure, STEM images of sample A were taken and the elemental mapping for Ga and Al were carried out on a selected area, as shown in Fig. 3. As we know, with HAADF-STEM method the image contrast is approximately proportional to the square of atomic number ($Z^2$). Thus, one

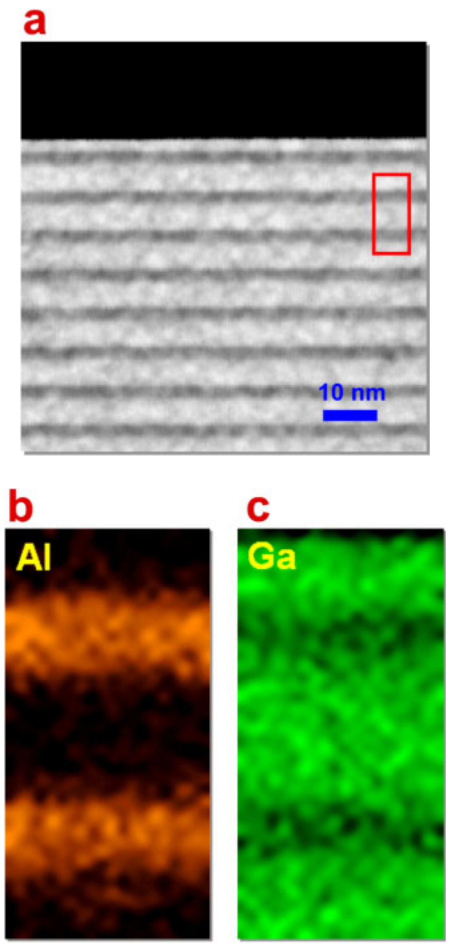

**Fig. 3.** (Color online) TEM images of GaN/AlGaN SLs structure of sample A. (a) Cross-sectional HAADF-STEM image of the cross section of AlGaN/GaN SLs, (b) and (c) elemental mapping of Al and Ga across the interface, respectively.

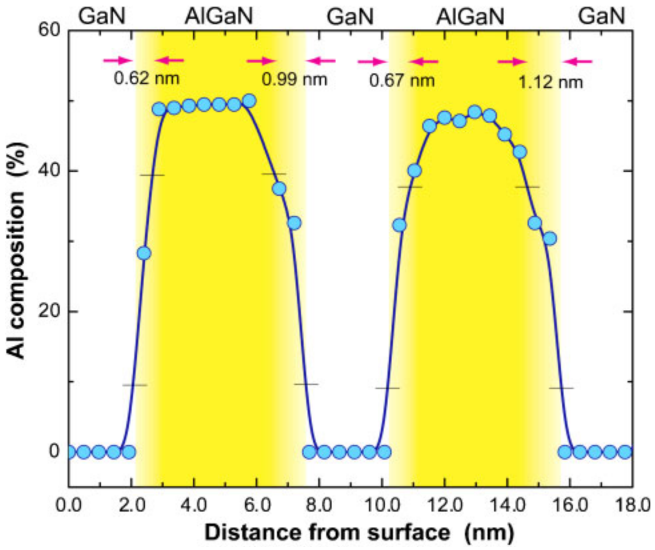

**Fig. 4.** (Color online) Al content profile across the interfaces of AlGaN/GaN SLs of sample A by AES. The diffusion depth of Al at interface is determined with the 20–80% criterion. An asymmetric diffusion at the upper and lower GaN interface is observed, which is strongly associated with interface condition.

can see that the bright and dark stripes in Fig. 3(a) directly correspond to the GaN wells and the AlGaN barriers, respectively. In detail, the thickness of the AlGaN and GaN layer measured from the image is about 3.2 and 4.7 nm, respectively. Since the well and barrier can be clearly distinguished, it seems that the heterointerfaces should be abrupt. However, when we look into the interfaces by elemental mapping [Figs. 3(b) and 3(c)], two interfaces can be identified and the GaN/AlGaN interface shows some diffusion of Al in the interfacial region. Due to the limitation of the lateral resolution of elemental mapping by EDX, the Al composition profile by AES was carried out to determine the diffusion depth and elemental abruptness of the heterointerfaces, as shown in Fig. 4. One can see that the transition of Al content at the GaN/AlGaN and AlGaN/GaN interfaces has an obviously asymmetric slope. By using the 20–80% criterion, it is determined that the diffusion depth at the AlGaN/GaN is about 0.62 nm, whereas that the GaN/AlGaN demonstrates a gradual transition from AlGaN to GaN where the diffusion of Al covers a depth of about 0.99 nm. As we know, Al adatoms are sticky to the surface and have much lower surface mobility than Ga adatoms. Hence, the AlGaN/GaN interface needs longer interruption time for the Al migration to approach a smoother interface. On the contrary, at the GaN/AlGaN interface the GaN layer turns to evaporate at high temperature during the growth interruption. This will lead to the interface roughening and after the interruption, the Al atoms easily undergo a downward diffusion into the GaN layer due to the steep gradient of Al content. Examination of sample B shows the similar phenomenon of considerable Al diffusion. This suggests that the Al diffusion at the GaN/AlGaN interface is a basic problem related directly to the high growth temperature and the Al concentration gradient across the interface, which is difficult to solve with conventional method. Note that the Al composition in the barrier appears a little bit higher than 0.4, which actually stems from the aforementioned etching effect on GaN at high temperature.

From the AES measurement in Fig. 4, an interesting behavior can be observed that the Al diffusion is significantly reduced when the Al composition becomes lower than 20%. This suggests a possible way to minimize the diffusion depth if a layer in lower Al content can be intentionally inserted to the interface. That inserted layer might block the diffusion of Al at high temperature to some degree. However, one may argue that since another layer with an Al content lower than that of the barrier is introduced to the interface, this additional layer itself will deteriorate the interfacial abruptness. In this concern, we propose a scheme with inserting a pair of layers to get the balance. This pair includes a ultra-thin low-Al-content layer acting as a blocking layer (B1) and another higher-Al-content layer (B2) playing a role of compensating the low Al content in B1, as schematically shown in Fig. 5(a). The B1 is able to block the Al diffusion from AlGaN into GaN, whereas an intentional diffusion from the B2 layer into B1 layer could increase the Al content in B1 and make the average composition equal to that of the AlGaN barrier. Based on this idea, the blocking-compensation pair layers were inserted into the GaN/AlGaN interface of sample C and the thickness of these ultra-thin blocking layers has been

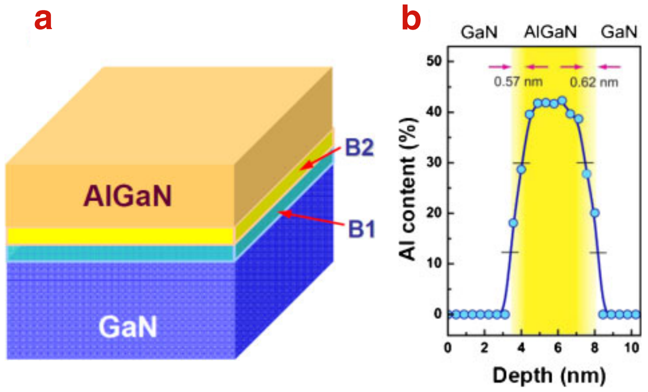

**Fig. 5.** (Color online) (a) Schematic of the blocking layer technique at the AlGaN/GaN interface which is introduced to cancel the asymmetric diffusion effect. The low-Al-content blocking (B1) is inserted to blocking the Al diffusion into the GaN layer, and the higher-Al-content compensation layer (B2) is set to increase the Al content in the B1 such that the average content of these two blocking layers can be equal to the AlGaN barrier. (b) Al content profile of AlGaN/GaN SLs of sample C with blocking layers, which shows symmetrical abrupt interfaces.

optimized. As a result, a thickness of 0.3 nm for each inserting layer is determined to be the best choice for achieving effective blocking. As shown in Fig. 5(b), the Al profile for the sample with blocking layers illustrates a symmetric and abrupt interface. The diffusion depth (abruptness) across the GaN/AlGaN interface has been improved to be about 0.62 nm, close to that at the AlGaN/GaN interface. Meanwhile, the XRD satellite of sample C shows the +3 order peak, confirming the abruptness of the interface as well. The room temperature PL was taken to characterize the emission intensity of the samples with and without blocking layers. It was found that the emission intensity of the SLs spectral peak could be enhanced by about 17%. The strong confinement for the carriers in a sharp quantum well is the reason for this emission efficiency improvement.

## 4. Conclusions

In conclusion, we proposed a simple blocking scheme to effectively enhance the abruptness of the AlGaN/GaN SLs. It was found that the Al diffusion depth at the upper and lower interfaces of the AlGaN barrier appears considerably asymmetric, which is about 0.62 and 0.96 nm, respectively. Such difference leads to the gradient interfacial region and the asymmetric quantum well shape. A pair of ultrathin blocking layers was introduced to the GaN/AlGaN interface to block the Al downward diffusion. The use of ultra-thin compensation layer is important for preserving the average Al content, equal to that of the AlGaN barrier. With the blocking treatment, the abruptness is improved and the light emission intensity from the SLs can be effectively enhanced. This scheme could provide effective blocking for high-temperature metal diffusion across the semiconductor heterointerface and improve the interfacial abruptness.

## Acknowledgments

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1) F. A. Ponce and D. P. Bour: Nature **386** (1997) 351.
2) J. W. Orton and C. T. Foxon: Rep. Prog. Phys. **61** (1998) 1.
3) S.-R. Jeon, S.-J. Lee, S. H. Jung, S. H. Lee, J. H. Baek, H. Jeong, O. H. Cha, E.-K. Suh, and M. S. Jeong: J. Phys. D **41** (2008) 132006.
4) S.-H. Han, D.-Y. Lee, S.-J. Lee, C.-Y. Cho, M.-K. Kwon, S. P. Lee, D. Y. Noh, D.-J. Kim, Y. C. Kim, and S.-J. Park: Appl. Phys. Lett. **94** (2009) 231123.
5) Z. Sitar, M. J. Paisley, B. Yan, J. Ruan, W. J. Choyke, and R. F. Davis: J. Vac. Sci. Technol. **8** (1990) 316.
6) K. Hoshino, T. Someya, K. Hirakawa, and Y. Arakawa: J. Cryst. Growth **237–239** (2002) 1163.
7) D. Fuhrmann, T. Retzlaff, U. Rossow, H. Bremers, A. Hangleiter, G. Ade, and P. Hinze: Appl. Phys. Lett. **88** (2006) 191108.
8) J. S. Cabalu, A. Bhattacharyya, C. Thomidis, I. Friel, T. D. Moustakas, C. J. Collins, and P. Komninou: J. Appl. Phys. **100** (2006) 104506.
9) A. Asgari, E. Ahmadi, and M. Kalafi: Microelectron. J. **40** (2009) 104.
10) E. Baumann, F. R. Giorgetta, D. Hofstetter, H. Lu, X. Chen, W. J. Schaff, L. F. Eastman, S. Golka, W. Schrenk, and G. Strasser: Appl. Phys. Lett. **87** (2005) 191102.
11) D. Hofstetter, E. Baumann, F. R. Giorgetta, F. Guillot, S. Leconte, and E. Monroy: Appl. Phys. Lett. **91** (2007) 131115.
12) A. Vardi, G. Bahir, F. Guillot, C. Bougerol, E. Monroy, S. E. Schacham, M. Tchernycheva, and F. H. Julien: Appl. Phys. Lett. **92** (2008) 011112.
13) L. Nevou, N. Kheirodin, M. Tchernycheva, L. Meignien, P. Crozat, A. Lupu, E. Warde, F. H. Julien, G. Pozzovivo, S. Golka, G. Strasser, F. Guillot, E. Monroy, T. Remmele, and M. Albrecht: Appl. Phys. Lett. **90** (2007) 223511.
14) Y.-T. Moon, D.-J. Kim, K.-M. Song, D.-W. Kim, M.-S. Yi, D.-Y. Noh, and S.-J. Park: J. Vac. Sci. Technol. B **18** (2000) 2631.
15) T. Shirasawa, N. Mochida, A. Inoue, T. Honda, T. Sakaguchi, F. Koyama, and K. Iga: J. Cryst. Growth **189–190** (1998) 124.
16) R. G. Banal, M. Funato, and Y. Kawakami: Phys. Status Solidi C **7** (2010) 2111.