Extremely high internal quantum efficiencies from AlGaN/AlN quantum wells emitting in the deep ultraviolet spectral region

Ryan G. Banal,[a)] Mitsuru Funato, and Yoichi Kawakami[b)]
Department of Electronic Science and Engineering, Kyoto University, Kyoto 615-8510, Japan

(Received 12 April 2011; accepted 13 June 2011; published online 6 July 2011)

Internal quantum efficiencies (IQEs) as high as 69% were realized at room temperature from AlGaN/AlN quantum wells (QWs) emitting at 247 nm grown by metalorganic vapor phase epitaxy. The extremely high IQEs were achieved by examining the source–supply sequence. QWs fabricated by a continuous source–supply method have longer emission wavelengths (λ) and higher IQEs compared to QWs fabricated by modified migration enhanced epitaxy (MMEE). MMEE is an alternating source–supply method where the NH_3 interruption promotes Ga evaporation. Thus, to obtain the same λ, MMEE requires a lower growth temperature than the continuous method, compromising the quality of the AlN and AlGaN layers as well as the IQE of QWs. © *2011 American Institute of Physics*. [doi:10.1063/1.3607306]

Due to intensive research efforts over the years, internal quantum efficiencies (IQEs) of AlGaN-based quantum wells (QWs) have improved and are catching up with their InGaN counterpart.[1] In fact, an IQE of 70% has been realized using AlGaN/AlGaN multiple QWs (MQWs) emitting at 280 nm at room temperature (RT).[2] However, the IQE tends to decrease at shorter emission wavelengths (λ). For example, the reported IQEs for AlGaN/AlN MQWs are 50% at 250 nm and 5% at 220 nm.[3] In addition, we have reported a 36% IQE (λ ~240 nm) from a 5 nm thick AlGaN/AlN MQW fabricated by modified migration enhanced epitaxy (MMEE),[4] a type of alternating source–supply method based on metalorganic vapor phase epitaxy (MOVPE). Reducing the QW thickness further improves the IQE to 57% (λ ~237 nm), thereby demonstrating a highly efficient electron-beam pumped DUV light source.[5]

The remarkable improvements in IQEs rely on the high-quality Al(Ga)N underlying epilayer achieved via various approaches, including alternating supply[6–11] and continuous supply[12] of group III and V precursors. Moreover, the QW active layer must be optimized for a number of factors such as growth temperature (T_g), Ga flow rate, and growth method. In principle, the optimal T_g of AlGaN with a given Al composition (x) is determined by the material properties such as cohesive energy and should be between those of GaN and AlN. In practice, however, the situation is not that simple because the aforementioned factors are closely correlated with each other. For example, a higher T_g requires a higher Ga flow rate to compensate for Ga evaporation due to its higher vapor pressure ($\sim 1.2 \times 10^{-1}$ Torr for Ga against $\sim 6.1 \times 10^{-3}$ Torr for Al at 1200 °C). In addition, for AlGaN/AlN QWs, their T_g requires a compromise between the AlGaN well and AlN barrier growths because either of them has to be grown at an un-optimized T_g.

In this Letter, we examine the appropriate growth conditions for QWs fabricated by MMEE[4,11] or the continuous source-supply method. Then their differences in emission wavelength and IQEs are investigated. These efforts realize AlGaN/AlN QWs with extremely high IQEs.

The samples were grown on (0001) sapphire substrates by MOVPE. The growth parameters are reported elsewhere.[4,11] The samples were composed of a ~600 nm thick AlN underlying layer and AlGaN/AlN single QWs (SQWs) or MQWs with a barrier and well thickness of ~14 nm and 1–2 nm, respectively. The MQW period was eight unless stated. The entire structure was fabricated by either MMEE[4,11] or the continuous method. As illustrated in Fig. 1, MMEE is characterized by an alternating supply of metalorganic precursors [trimethylaluminum (TMA) and trimethylgallium (TMGa)] and NH_3. To increase Ga incorporation in the AlGaN wells, T_g was decreased from the optimum T_g for AlN (~1200 °C) towards that for GaN (~1050 °C). The IQE was estimated from the ratio of integrated photoluminescence (PL) intensities measured at 8.5 K and RT. The excitation source was an ArF laser (193 nm) with a power density of 100 kW/cm^2. IQEs estimated in this manner depends on the excitation power,[2] and 100 kW/cm^2 provides almost maximum estimates of IQE.[13]

Figure 2 shows the RT PL spectra of AlGaN/AlN SQWs fabricated by these two methods at the optimum T_g for AlN. Although they were grown under identical conditions, except for the flow sequence of the precursors, the PL peak wavelength obtained from MMEE (~215 nm) is much shorter than that obtained from the continuous method (~233 nm).

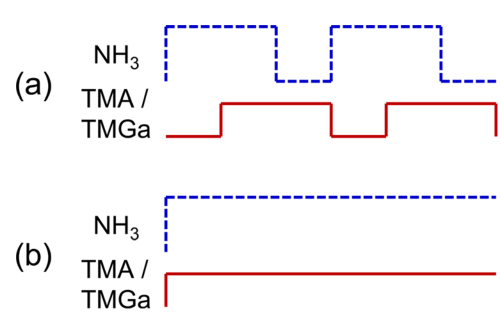

FIG. 1. (Color online) Sequence profile of NH_3 and metalorganic sources (TMA and TMGa) for (a) MMEE and (b) continuous methods.

[a)]Electronic mail: ryan.banal@optomater.kuee.kyoto-u.ac.jp.
[b)]Electronic mail: kawakami@kuee.kyoto-u.ac.jp.

0003-6951/2011/99(1)/011902/3/$30.00

© 2011 American Institute of Physics

Downloaded 24 Aug 2011 to 130.54.110.73. Redistribution subject to AIP license or copyright; see http://apl.aip.org/about/rights_and_permissions

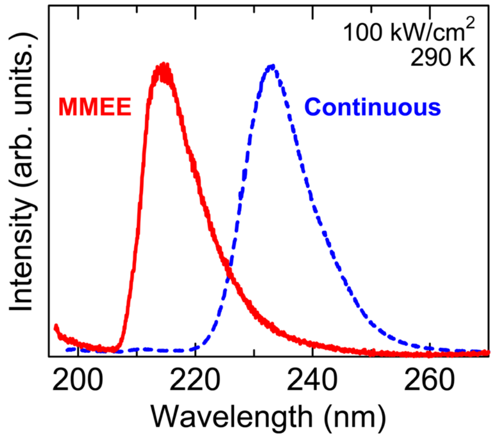

FIG. 2. (Color online) RT PL spectra of AlGaN/AlN SQWs fabricated by MMEE and continuous method under the same growth conditions except for the source-supply sequence.

Difference in QW thickness between the growth methods cannot be the reason for this after ensuring similar thickness by considering the growth rates and adjusting the growth period accordingly. Rather, this result clearly indicates that Ga incorporation in the well layer differs between the two methods. As depicted in Fig. 1, in every 3 s period of MMEE, NH_3 is interrupted for 1 s. During this interruption, Ga desorption is enhanced because Ga has a higher vapor pressure than Al. Thus, when desiring the same λ, MMEE requires a lower T_g than the continuous method to prevent Ga desorption.

Next, a series of QWs emitting at different λ were fabricated by both methods. As expected, Ga incorporation increases with decreasing T_g, but for the continuous method, QWs emitting at λ shorter than 230 nm had to be grown by reducing the TMGa flow rates. Figure 3 summarizes the experimentally estimated IQEs as well as the calculated square overlap of the electron and hole wavefunctions as functions of the peak emission wavelength at RT. The assumptions were made in the calculation: fully coherent structure, presence of polarization-induced field, and exclusion of excitonic effect and localization. QWs fabricated by the continuous method have higher IQEs compared to those fabricated by MMEE. In fact, at a similar λ of ~240 nm, the highest IQE obtained from MMEE is 57%, whereas the highest for the continuous method is 69%. Hence, the factors causing the IQE differences were investigated.

One possible cause is carrier localization which enhances IQEs in AlGaN[2,3] and InGaN.[14,15] To examine the difference in the degree of localization due to the growth method, the PL line widths with λ at 8.5 K were compared in Fig. 4. Both methods provide comparable line widths, which broaden inhomogeneously. Therefore, both methods yield similar degrees of localization and cannot explain the IQE differences.

Another is crystalline quality. However, studies from numerous QWs grown by either method do not show a clear correlation between x-ray diffraction line widths and IQE. For example, one MQW with an IQE of 48% exhibits ω-scan line widths of 200 arcsec for the (0002) plane and 1400 arcsec for the $(10\bar{1}2)$ plane, while another MQW with an IQE of 31% shows better respective line widths of 80 and 1250 arcsec. Therefore, the microscopic screw and edge dislocation densities are not likely causes for the IQE difference.

Although the experimental evidence is unclear, we believe that nanoscopic defects and/or impurities are responsible for the observed IQE difference. Note that T_g drastically differs for the two growth methods; QWs with λ ~240 nm can be grown at ~1165 °C by the continuous method, whereas the MMEE requires ~1080 °C. Moreover, because QWs are grown below the optimum T_g for AlN, MMEE at even much lower T_g may induce unexpected impurities, point defects and/or interstitials not only into the AlN barrier,[16] but more importantly into the AlGaN QWs. This explains why the continuous method always gives a higher IQE than MMEE.

Let us discuss the emission wavelength dependence of IQE shown in Fig. 3. Regardless of the growth method, the IQE initially increases with λ, that is, with the Ga composition. One reason for this may be stronger quantum confinement. To quantify this effect, Fig. 3 also plots the square overlap of the electron and hole wavefunctions. In fact, this effect strengthens the IQE, particularly at shorter λ. In addition, as shown in Fig. 4, the localization degree increases with λ, which may also contribute to the observed wavelength dependence of IQE.

After reaching the maximum, the IQE decreases with λ. As previously mentioned, a longer λ requires a lower T_g,

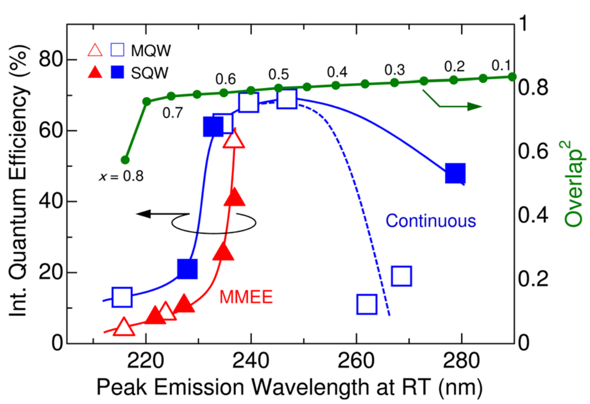

FIG. 3. (Color online) Summary of IQEs with emission wavelength for QWs grown by MMEE or continuous method. Calculated square overlap is also shown. Numbers in italics are Al compositions.

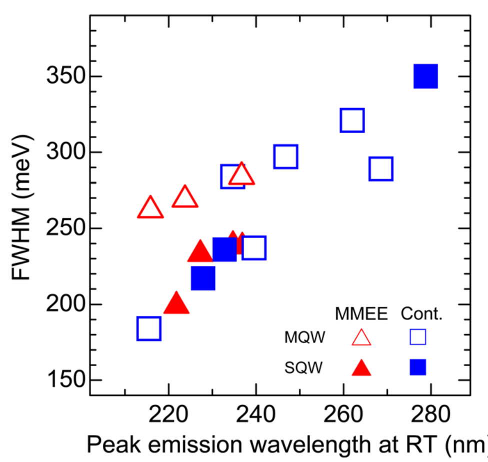

FIG. 4. (Color online) Variation in the PL line width of QWs at 8.5 K with emission wavelength at RT.

Downloaded 24 Aug 2011 to 130.54.110.73. Redistribution subject to AIP license or copyright; see http://apl.aip.org/about/rights_and_permissions

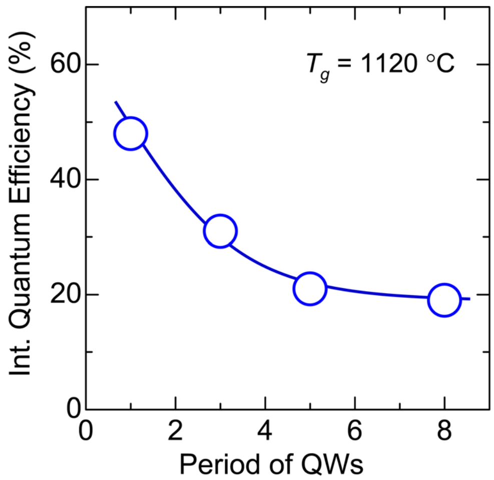

FIG. 5. (Color online) Variation of IQE due to QW period in 1.5 nm thick $Al_{0.50}Ga_{0.50}N$/AlN QWs grown by the continuous method.

which may degrade the QW quality. Additionally, because the lattice mismatch between AlGaN and AlN becomes larger at longer λ, strain-related phenomena should be considered. Although the piezoelectric polarization enhances the internal electric field, the calculated square overlap does not decrease with λ (Fig. 3). This can be explained by considering that the internal electric field, which is strengthened at longer λ, is reconciled with the increased carrier confinement. Another possible explanation for the IQE decrease at longer λ is lattice relaxation. To confirm this effect, Fig. 3 compares a SQW (solid line) and 8-period MQWs (broken line). A sharper decrease of IQEs from MQWs suggests the lattice relaxation.

To more thoroughly examine the effect of AlGaN thickness, we fabricated a series of 1.5 nm thick $Al_{0.50}Ga_{0.50}N$/AlN QWs with various periods by the continuous method. As plotted in Fig. 5, the IQE decreases as the period increases. An especially sharp drop observed between periods one and three indicates that the lattice relaxation occurs during these periods. Note that periods one and three correspond to AlGaN thicknesses of 1.5 and 4.5 nm, respectively. Hence, the critical thickness (h_c) for AlGaN/AlN QW was calculated using Matthews–Blakeslee (M–B)[17] and Fischer[18] models assuming a (0001) slip plane, 90° dislocation with a Burgers vector of $1/3[11\ \bar{2}\ 0]$, and a Poisson's ratio of 0.38. For the QW with $x \sim 0.50$, the estimated h_c is ~2 nm from the M–B model and ~16 nm from the Fischer model. Therefore, all the SQWs with x above 0.50 are fully strained, and the experimentally observed lattice relaxation between 1.5 and 4.5 nm suggests that the M–B model can better reproduce the situation in AlGaN/AlN QWs. To avoid the lattice relaxation and to obtain a high IQE at longer λ, an alternative to the AlN underlying epilayer would be a ternary AlGaN layer.[2]

The results imply a compromise between the growth methods; although MMEE achieves an AlN epilayer with a superior crystal quality,[4,11] the continuous method is a better choice for growing AlGaN QWs. (Although high-quality AlGaN wells can be obtained from MMEE, they are limited to the shorter λ region.) We are in the process of fabricating AlGaN QWs by the continuous method on MMEE-AlN to verify this hypothesis.

[1] Y. Narukawa, M. Ichikawa, D. Sanga, M. Sano, and T. Mukai, J. Phys. D: Appl. Phys. **43**, 354002 (2010).
[2] M. Shatalov, J. Yang, W. Sun, R. Kennedy, R. Gaska, K. Liu, M. Shur, and G. Tamulatitis, J. Appl. Phys. **105**, 073103 (2009).
[3] A. Bhattacharyya, T. D. Moustakas, L. Zhou, D. J. Smith, and W. Hug, Appl. Phys. Lett. **94**, 181907 (2009).
[4] R. G. Banal, M. Funato, and Y. Kawakami, Phys. Status Solidi C **7**, 2111 (2010).
[5] T. Oto, R. G. Banal, K. Kataoka, M. Funato, and Y. Kawakami, Nature Photon. **4**, 767 (2010).
[6] J. P. Zhang, M. A. Khan, W. H. Sun, H. M. Wang, C. Q. Chen, Q. Fareed, E. Kuokstis, and J. W. Yang, Appl. Phys. Lett. **81**, 4392 (2002).
[7] M. Hiroki and N. Kobayashi, Jpn. J. Appl. Phys., Part 1 **42**, 2305 (2003).
[8] V. Adivarahan, W. H. Sun, A. Chitnis, M. Shatalov, S. Wu, H. P. Maruska, and M. A. Khan, Appl. Phys. Lett. **85**, 2175 (2004).
[9] M. Takeuchi, H. Shimizu, R. Kajitani, K. Kawasaki, Y. Kumagai, A. Koukitu, and Y. Aoyagi, J. Cryst. Growth **298**, 336 (2007).
[10] H. Hirayama, T. Yatabe, N. Noguchi, T. Ohashi, and N. Kamata, Appl. Phys. Lett. **91**, 071901 (2007).
[11] R. G. Banal, M. Funato, and Y. Kawakami, Appl. Phys. Lett. **92**, 241905 (2008).
[12] K. Balakrishnan, A. Bandoh, M. Iwaya, S. Kamiyama, H. Amano, and I. Akasaki, Jpn. J. Appl. Phys. **46**, L307 (2007).
[13] Y. Iwata, T. Oto, A. Kaneta, R. G. Banal, M. Funato, and Y. Kawakami, Phys. Status Solidi C (in press).
[14] Y. Narukawa, Y. Kawakami, M. Funato, Sz. Fujita, Sg. Fujita, and S. Nakamura, Appl. Phys. Lett. **70**, 981 (1996).
[15] A. Kaneta, M. Funato, and Y. Kawakami, Phys. Rev. B **78**, 125317 (2008).
[16] S. F. Chichibu, T. Onuma, K. Hazu, and A. Uedono, Appl. Phys. Lett. **97**, 201904 (2010).
[17] J. W. Matthews and A. E. Blakeslee, J. Cryst. Growth **27**, 118 (1974).
[18] A. Fischer, H. Kuhne, and H. Richter, Phys. Rev. Lett. **73**, 2712 (1994).

Downloaded 24 Aug 2011 to 130.54.110.73. Redistribution subject to AIP license or copyright; see http://apl.aip.org/about/rights_and_permissions