12-24-2001

Localization of Carriers and Polarization Effects in Quaternary AlInGaN Multiple Quantum Wells

E. Kuokstis

J. Zhang

M.-Y. Ryu

J. W. Yang

Grigory Simin
University of South Carolina - Columbia, simin@engr.sc.edu

See next page for additional authors

Publication Info

Published in *Applied Physics Letters*, Volume 79, Issue 26, 2001, pages 4375-4377.

Kuokstis, E., Zhang, J., Ryu, M.-Y., Yang, J. W., Simin, G., Khan, M. A., Gaska, R., & Shur, M. S. (24 December 2001). Localization of Carriers and Polarization Effects in Quaternary AlInGaN Multiple Quantum Wells. Applied Physics Letters, 79 (26), 4375-4377. http://dx.doi.org/10.1063/1.1429753

AIP Applied Physics Letters

Localization of carriers and polarization effects in quaternary AlInGaN multiple quantum wells

E. Kuokstis, J. Zhang, M.-Y. Ryu, J. W. Yang, G. Simin, M. Asif Khan, R. Gaska, and M. S. Shur

Citation: Applied Physics Letters **79**, 4375 (2001); doi: 10.1063/1.1429753
View online: http://dx.doi.org/10.1063/1.1429753
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/79/26?ver=pdfcov
Published by the AIP Publishing

Localization of carriers and polarization effects in quaternary AlInGaN multiple quantum wells

E. Kuokstis,[a] J. Zhang, M.-Y. Ryu, J. W. Yang, G. Simin, and M. Asif Khan
Department of Electrical Eengineering, University of South Carolina, Columbia, South Carolina 29208

R. Gaska
Sensor Electronic Technology, Inc., 21 Cavalier Way, Latham, New York 12110

M. S. Shur
ECSE, Rensselaer Polytechnic Institute, Troy, New York 12180 and Sensor Electronic Technology, Inc., 21 Cavalier Way, Latham, New York 12110

(Received 20 August 2001; accepted for publication 1 November 2001)

We report on observing a long-wavelength band in low-temperature photoluminescence (PL) spectrum of quaternary $Al_{0.22}In_{0.02}Ga_{0.76}N/Al_{0.38}In_{0.01}Ga_{0.61}N$ multiple quantum wells (MQWs), which were grown over sapphire substrates by a pulsed atomic-layer epitaxy technique. By comparing the excitation-power density and temperature dependence of the PL spectra of MQWs and bulk quaternary AlInGaN layers, we show this emission band to arise from the carrier and/or exciton localization at the quantum well interface disorders. PL data for other radiative transitions in MQWs indicate that excitation-dependent spectra position is determined by screening of the built-in electric field. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1429753]

III nitrides are currently under extensive investigation due to their ability to emit light over most of visible and near UV spectrum. To date, light emitting diodes or laser diodes are mainly based on multiple quantum wells (MQWs) of binary GaN and ternary InGaN or AlGaN layers.[1,2] Recently we have shown the advantages of using quaternary AlInGaN layers for the fabrication of high quality quantum structures[3–5] with strong UV emission at room temperature.[6] Due to an independent tunability of band gap and lattice constant the quaternary AlInGaN alloys provide an excellent vehicle for band engineering[7–9] and the investigation of strain and built-in electric field effects in quantum wells.[10]

In this letter, we present a systematic study of the photoluminescence (PL) of quaternary AlInGaN MQW structures with a band to band emission at 320 nm. The PL study was carried out at temperatures from 7 to 300 K and excitation pump power densities were varied over five orders. Emission properties of quaternary AlInGaN/AlInGaN MQWs and thick AlInGaN epilayers were also compared in order to establish the mechanisms responsible for the observed emission bands.

The structures for this study were grown over basal plane sapphire substrates using a pulsed atomic layer epitaxy (PALE) technique.[11] This growth method allows the deposition of high-quality III–N layers at temperatures approximately 200–300 °C lower than that required for conventional metalorganic chemical vapor deposition (MOCVD).[12,13] This allowed for increased In incorporation and in turn improved the quantum well interfaces and the structural properties of the lattice.[5] The studied structures were comprised of a 1.5-μm-thick intrinsic *i*-GaN buffer layer followed by either a 0.1-μm-thick "bulk" $Al_xIn_yGa_{1-x-y}N$ layer or an $Al_xIn_yGa_{1-x-y}N$–$Al_xIn_yGa_{1-x-y}N$ MQW. Conventional MOCVD was used for the buffer GaN growth while the quaternary layers were deposited by the PALE approach. Al molar fraction in the quantum wells and barrier layers was close to 20% and 40%, respectively. Indium content in both wells and barriers did not exceed 2%. The MQW structures consisted of four 4-nm-thick wells separated by 5-nm-thick barriers.

Samples were mounted on the cold finger of a closed-cycle He cryostat and the PL spectra were then measured in a wide temperature range (7–300 K) using pulsed excimer laser excitation ($\lambda = 193$ nm, $\tau = 8$ ns). The laser beam was focused on the surface of the sample to a spot of about 0.1 mm diameter. A maximum pump power density of ~2 MW/cm^2 could thus be reached. Luminescence was measured in a backscattering geometry using a SPEX550 monochromator with a UV-enhanced charge coupled device array.

The low-temperature (7 K) PL spectra of the AlInGaN MQW and bulk epilayer with composition identical to the well material at different excitation power densities are shown in Figs. 1(a) and 1(b), respectively. For the epilayers only one band with a maximum at 3.780 eV is observed [Fig. 1(a)]. Its position remains unchanged up to an excitation power density of about 5 kW/cm^2. A further increase in excitation leads to a small blueshift (~14 meV). This is also illustrated in Fig. 2 by open circles.

PL spectra of MQWs, however, have quite different features and behavior as a function of excitation power density. As seen from Fig. 1(b) they consist of two bands. At an excitation power density below ~3 kW/cm^2 a long-wavelength (low-energy) band dominates. At higher excitations a new short-wavelength (high-energy) band appears. It dominates the spectrum until ~2 MW/cm^2 which is the maximum possible laser power density for our experimental setup. The position of both these MQW PL bands shifts to-

[a]Author to whom correspondence should be addressed; on leave from Department of Semiconductor Physics, Vilnius University, Sauletekio al. 9, Vilnius, 2040, Lithuania; electronic mail: koukstis@engr.sc.edu

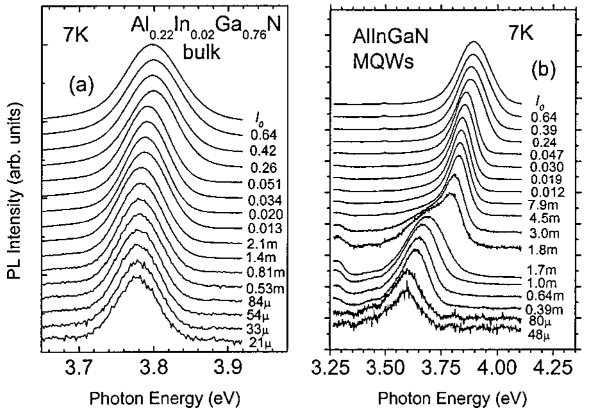

FIG. 1. $Al_{0.22}In_{0.02}Ga_{0.76}N$ compound epilayer (well material) (a) and $Al_{0.22}In_{0.02}Ga_{0.76}N/Al_{0.38}In_{0.01}Ga_{0.61}N$ MQW (b) spectra at 7 K under different excitations. The top spectra correspond to the maximum excimer laser power density $I_0 = 2$ MW/cm^2. The figures at lower curves indicate the fraction of excitation power density with respect to I_0; "*m*" stands for 10^{-3}, and "μ" stands for 10^{-6}.

wards higher energies with increasing excitation power. The total shift is around 300 meV when excitation is increased from 40 W/cm^2 to 2 MW/cm^2. This is shown in Fig. 2 where open and filled squares correspond to the long and the short-wavelength bands, respectively.

We believe the observed differences in the PL properties of quaternary MQWs and epilayers arise from the quantum origin of the structures. For MQWs internal electric field from the piezoelectric[14] and spontaneous polarizations,[10,15] as well as electron-hole pair (exciton) localization at the wells of random potential induced by alloy disorder[16] or interface disorders in quantum structures,[17,18] should be taken into account. Note that both localization and electric fields lead to blueshift with increased excitation.[19]

To determine the mechanism for the observed PL of our MQW structure we first analyzed its built-in electric field. For the calculation of this internal field we used parameters of AlN, GaN, and InN and linearly extrapolated them according to their molar fraction in the quaternary alloy. As the GaN buffer layer is thick ($\sim$1.5 μm), we assume it to be

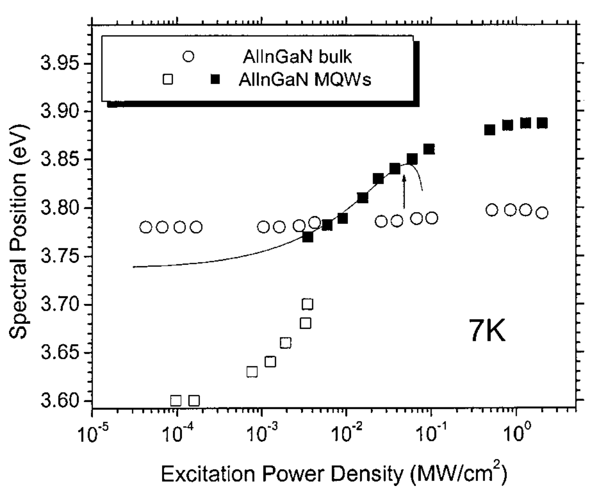

FIG. 2. PL maximum spectral position as a function on excimer laser excitation power density in AlInGaN well material epilayer (open circles) and MQWs (squares). Open squares correspond to long-wave band and filled squares correspond to short-wave band. Solid curve is a theoretical dependence.

completely relaxed. Therefore the thin AlInGaN quantum structures grown on top of it undergo tensile in-plane strain due to lattice mismatch and they therefore remain in a pseudomorphic state. Our calculations have shown that barriers and wells undergo the tension of 0.815% and 0.314%, respectively. Surface density of piezoelectric charge induced by this in-plane mismatch at barrier and well interfaces was calculated[14] -0.0484 C/m^2 for well and -0.0134 C/m^2 for barrier. Meanwhile, spontaneous polarization charge density according to Ref. 13 was found to be -0.041 and -0.049 C/m^2 for wells and barriers, respectively. The resultant built-in electric field in the wells of our MQWs thus was calculated to be 1.2 MV/cm. We also estimated that 50% of the total electric field was due to piezoelectric effect and the rest (50%) was caused by spontaneous polarization.

In order to compare our experimental data with the theoretical model we have calculated the variation of luminescence maximum energy position with excitation power using a triangular potential well[20] resulting from the built-in electric well field. We took into account the screening of electric field by photoexcited carriers[21] whose number was estimated from the laser excitation power density after assuming that square recombination of carriers predominates.[22] The results of these theoretical estimations are shown in Fig. 2 as the solid curve. For excitation power density up to 50 kW/cm^2 they agree well with the experimental blueshift data of Fig. 2. At excitation power densities higher than 50 kW/cm^2 (arrow in Fig. 2) complications arise due to a very high density of nonequilibrium carriers. This requires inclusion of effects such as the heating of quasiparticle systems,[23] lifetime reduction due to stimulated transitions,[24] or Auger recombination. Note that at these excitations the triangular potential well model breaks down.

A clearly resolved separate band at low excitation, as well as our theoretical estimations of the PL peak position behavior (for a quaternary MQWs) suggest the low-energy PL band to originate from a different radiative recombination mechanism. Indeed, the energy position of this band is well below the value predicted for transitions between levels in quantum wells (compare open squares and solid curve in Fig. 2). Increased excitation power leads to saturation and a simultaneous blueshift (see Fig. 1 and Fig. 2). At higher temperatures the band is not clearly resolved. Figure 3 illustrates the change of energy position with temperature for this and the high-energy band at higher excitations. With increased temperatures, the low-energy PL band undergoes a slight blueshift. This is opposite to the behavior of the high-energy band which shows a redshift similar to bulk material.

The most probable mechanism for the origin of this low-energy band is radiative recombination of localized excitons or carriers. Indeed, a distinctive feature of solid-solution quantum wells is that the fluctuation effects occur even in systems with atomic substitution in the cationic sublattice.[16] At low temperatures the fluctuation states do not play a significant role in the recombination processes in three-dimensional (bulk) materials.[18] However, in quantum wells carriers and excitons can always be localized by interface disorders. These can occur from the local height fluctuations such as island-like structures having a height of 1 ML.[17] For such a case a density of state tail occurs below the forbidden

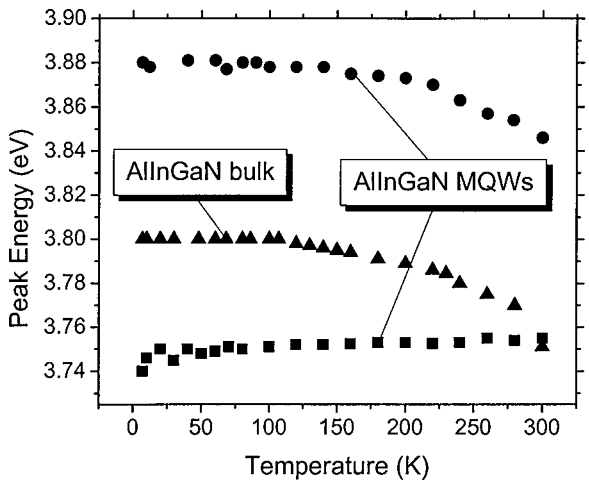

FIG. 3. AlInGaN compound epilayer (triangles) and AlInGaN MQW (circles and squares) PL maximum position dependence on temperature. Circles and triangles correspond to excimer laser excitation power density about 1 MW/cm^2, whereas squares correspond to about 3 kW/cm^2.

gap. This should give rise to an emission band redshifted with respect to absorption edge. An increased excitation then leads to the filling of band tail states and hence a blueshift of the emission band. Such a band should also vanish with increasing temperature. A low-temperature PL band with similar features was also observed for disordered II–VI solid solutions[16] and was assigned to localized band-tail states. We therefore conclude that the emission band observed in the low-temperature PL of quaternary MQWs arises from electron-hole pairs localized at quantum well-barrier interface disorders.

In summary, we report on low-temperature PL in quaternary $Al_{0.38}In_{0.01}Ga_{0.61}N/Al_{0.22}In_{0.02}Ga_{0.76}N$ multiple quantum wells (MQWs) and bulk epilayers grown by a pulsed atomic-layer epitaxy technique. A low-energy PL emission band is observed. Comparison of PL spectra of bulk epilayers and MQWs under different excitaion power densities and temperatures from 7 to 300 K shows this PL band to be from localized electron-hole pairs (presumably as excitons) at quantum-well interface. The strong blueshift of PL maximum with excitation (~300 meV) is explained by a combination of the localized states filling and screening the built-in electric field from the piezo and spontaneous polarizations.

This work at USC was supported by the Ballistic Missile Defense Organization (BMDO) under U.S. Army SMDC Contract No. DASG60-98-1-0004, monitored by Terry Bauer, Dr. Brian Strickland, and Dr. Kepi Wu. The work at SET, Inc. was supported by the Office of Naval Research under the Small Business Technology Transfer Program monitored by Dr. Y.-S. Park.

[1] S. J. Pearton, J. C. Zolper, R. J. Shul, and F. Ren, J. Appl. Phys. **86**, 1 (1999).
[2] S. Nakamura and G. Fasol, *The Blue Laser Diode* (Springer, Berlin, 1997).
[3] M. A. Khan, J. W. Yang, G. Simin, R. Gaska, M. S. Shur, G. Tamulaitis, A. Zukauskas, D. J. Smith, D. Chandrasekhar, and R. Bicknell-Tassius, Appl. Phys. Lett. **76**, 1161 (2000).
[4] G. Tamulaitis, K. Kazlauskas, S. Jursenas, A. Zukauskas, M. A. Khan, T. W. Yang, J. Zhang, G. Simin, M. S. Shur, and R. Gaska, Appl. Phys. Lett. **77**, 2136 (2000).
[5] J. Zhang, J. W. Yang, G. Simin, M. Shatalov, M. A. Khan, M. S. Shur, and R. Gaska, Appl. Phys. Lett. **77**, 2668 (2000).
[6] E. Kuokstis, J. Zhang, J. W. Yang, G. Simin, M. A. Khan, R. Gaska and M. S. Shur, Appl. Phys. Lett. (to be published).
[7] T. Matsuoka, N. Yoshimoto, T. Sasaki, and A. Katsui, J. Electron. Mater. **21**, 157 (1992).
[8] J. Han, J. J. Figiel, G. A. Petersen, S. M. Myers, M. H. Crawford, and M. A. Banas, Jpn. J. Appl. Phys., Part 1 **39**, 2372 (2000).
[9] J. Li, K. B. Nam, K. H. Kim, J. Y. Lin, and H. X. Jiang, Appl. Phys. Lett. **78**, 61 (2001).
[10] A. Zaroddu, F. Bernardini, and P. Ruggerone, Phys. Rev. B **64**, 045208 (2001).
[11] M. A. Khan, R. A. Skogman, J. M. Van Hove, D. T. Olson, and N. Kuzina, Appl. Phys. Lett. **60**, 1366 (1992).
[12] M. A. Khan, N. Kuzina, R. A. Skogman, D. T. Olson, M. MacMillan, and W. J. Choyke, Appl. Phys. Lett. **61**, 2539 (1992).
[13] M. A. Khan, J. N. Kuzina, D. T. Olson, T. George, and W. T. Pike, Appl. Phys. Lett. **63**, 3470 (1993).
[14] T. Takeuchi, S. Sota, M. Katsuragawa, M. Komori, H. Takeuchi, H. Amano, and I. Akasaki, Jpn. J. Appl. Phys., Part 2 **36**, L382 (1997).
[15] F. Bernardini, V. Fiorentini, and D. Vanderbilt, Phys. Rev. B **56**, R10024 (1997).
[16] A. Klochikin, A. Reznitsky, S. Permogorov, T. Breitkopf, M. Grü, M. Hetterich, C. Klingshirn, V. Lysenko, W. Langbein, and J. M. Hvam, Phys. Rev. B **59**, 12947 (1999).
[17] T. Takahara, Phys. Rev. B **32**, 7013 (1985).
[18] A. Klochikhin, A. Reznitskiĭ, L. Tanishev, S. Permogorov, S. Ivanov, S. Sorokin, Kh. Mumanis, R. Seisyan, and C. Klingshirn, JETP Lett. **71**, 242 (2000).
[19] S. Chichibu, T. Sota, K. Wada, and S. Nakamura, J. Vac. Sci. Technol. B **16**, 2204 (1998).
[20] C. Weisbuch and B. Vinter, *Quantum Semiconductor Structures* (Academic, London, 1991).
[21] R. Cingolani, A. Botcharev, H. Tang, H. Markoç, G. Traetta, G. Coli, M. Lomascolo, A. Di Carlo, F. Della Sala, and P. Lugli, Phys. Rev. B **61**, 2711 (2000).
[22] A. Dmitriev and A. Oruzheinikov, J. Appl. Phys. **86**, 3241 (1999).
[23] S. Juršenas, G. Kurilčik, and A. Žukauskas, Phys. Rev. B **54**, 16706 (1996).
[24] R. Klann, O. Brandt, H. Yang, H. T. Grahn, and K. H. Ploog, Appl. Phys. Lett. **70**, 1076 (1997).