phys. stat. sol. (c) **2**, No. 7, 2779–2782 (2005) / **DOI** 10.1002/pssc.200461498

Photoluminescence study of MBE grown InGaN with intentional indium segregation

Maurice C. Cheung[*1], **Gon Namkoong**[2], **Fei Chen**[1], **Madalina Furis**[1], **Haridas E. Pudavar**[3], **Alexander N. Cartwright**[1], and **W. Alan Doolittle**[2]

[1] Department of Electrical Engineering, University at Buffalo, State University of New York, Buffalo, NY 14260, USA
[2] School of Electrical and Computer Engineering, Georgia Institute of Technology, Atlanta, GA 30332, USA
[3] Department of Chemistry, University at Buffalo, State University of New York, Buffalo, NY 14260, USA

Received 13 July 2004, revised 17 August 2004, accepted 24 January 2005
Published online 17 March 2005

PACS 78.47.+p, 78.55.Cr, 81.15.Hi

Proper control of MBE growth conditions has yielded an $In_{0.13}Ga_{0.87}N$ thin film sample with emission consistent with In-segregation. The photoluminescence (PL) from this epilayer showed multiple emission components. Moreover, temperature and power dependent studies of the PL demonstrated that two of the components were excitonic in nature and consistent with indium phase separation. At 15 K, time resolved PL showed a non-exponential PL decay that was well fitted with the stretched exponential solution expected for disordered systems. Consistent with the assumed carrier hopping mechanism of this model, the effective lifetime, τ, and the stretched exponential parameter, β, decrease with increasing emission energy. Finally, room temperature micro-PL using a confocal microscope showed spatial clustering of low energy emission.

© 2005 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

1 Introduction InGaN-based emitters are widely used for commercial high-brightness light emitting diodes (LEDs) from UV to amber [1]. The improvement of efficiency in these metal organic chemical vapor deposition (MOCVD) grown LED devices due to In-segregation has been well documented [2–5]. The current theory suggests that the In-rich regions existing inside InGaN act as localization centers that trap the carriers injected into the system. The probability of non-radiative recombination for a carrier localized in a potential fluctuation is significantly reduced [2, 3] and the excitons formed in the vicinity of the localization centers have greater oscillator strength [2]. As a result, the optical efficiency of the MOCVD grown LED is greatly improved. However, research is still required to control In-segregation in InGaN grown by molecular beam epitaxy (MBE).

It should be pointed out that the emission efficiency depends not only on the In-segregation but also on the strain induced piezoelectric fields present in the multiple quantum well (MQW) heterostructures that are generally used for commercial devices [6–10]. In order to eliminate this field effect when studying In-segregation, we have turned to the study of In-segregation of a MBE grown thin film InGaN sample. Using the precise growth control of MBE, we have intentionally grown samples that exhibit In-segregation.

In this paper, we present results of photoluminescence (PL), time resolved PL (TRPL), and spatially resolved PL measurements on an InGaN thin film sample that was intentionally grown with In-segregation. These results are consistent with localized emission and In-segregation.

[*] Corresponding author: e-mail: mccheung@eng.buffalo.edu, Phone: 001-716-645-3123, Fax: 001-716-645-3656

© 2005 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

2 Experiments The thin film sample used in this study is a MBE grown Si:InGaN with a nominal In mole fraction of 13 % and an epilayer thickness of 300 nm. The epilayer is grown on a multi-layered buffer deposited on a nitrated sapphire substrate. In the order of growth, the first buffer layer is a 30 nm GaN layer, the second layer is a 100 nm thick AlN and the third layer is a 500 nm Si:GaN.

The TRPL measurements were conducted at 15K. The 200 fs pulsed excitation (pump) was produced by a regenerative amplifier (Coherent: RegA) seeded by a Ti:Sapphire laser at 800 nm. An optical parametric amplifier (Coherent: OPA at 660 nm) and subsequent frequency doubling using a BBO crystal provided the up-conversion of the RegA pulses to a wavelength of 330 nm. The back-scattered light from the sample was collected through conventional optics, and spectrally and temporally resolved by a Chromex 250IS monochromator and a Hamamatsu C4334 streak camera with a typical temporal jitter of 50ps. Continuous wave (CW) PL was measured from 15K to 300K using the same detection system with a Coherent Sabre Ar^+ ion laser tuned to 351nm as the CW pump source. In this case, the streak camera was operated in focus mode to obtain the time-integrated PL. Temperature control was maintained by mounting the sample in a cryostat that was cooled by a closed cycle refrigerator.

Spatially resolved PL of the sample was obtained using a confocal microscope system with a lateral resolution of 200nm. In the PL measurement, a 457nm (2.71 eV) frequency doubled output beam of a Ti:sapphire laser was used as the pump, and it's beam was focused on the sample by the confocal microscope. For spectrally analyzing the PL, a computer-controlled spectrometer equipped with a CCD camera was connected to the microscope optical output through an optical fiber. When collecting the image of the luminescence, a long-pass filter with cut-off at 510 nm (2.43 eV) was used to remove the backscattering pump signal.

Table 1 Locations of the three Gaussian peaks fitted to the PL at 15 K and 300 K.

T (K)	Peak A	Peak B	Peak C
15	2.60 eV	2.72 eV	2.85 eV
300	2.59 eV	2.65 eV	2.83 eV

3 Results and discussion The sample has a broad PL emission, and with careful examination of the PL under CW excitation, we found that it is composed of three distinct Gaussian peaks labelled A, B and C respectively, as shown in Fig. 1(a). Their peak positions at 15 K and 300 K are listed in Table 1. The intensity, I, of each peak decreases with increasing temperature according to the Arrhenius dissociation model, and their respective Arrhenius plots are shown in Fig. 1(b). It was found that Peak A has one dissociation channel, while Peak B and Peak C have two. The estimated E_a of Peak A was 110 meV; the estimated values of E_a for the two channels in Peak B were 27 meV and 107 meV; and the estimated values of E_a for the two channels in Peak C were 26 meV and 125 meV. Note that the lower value of E_a for Peak B and Peak C match the accepted values of the exciton binding energy in InGaN [11], suggest

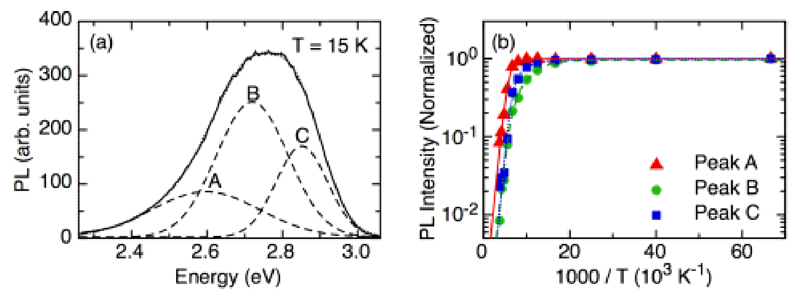

Fig. 1 (a) PL of the sample under CW excitation at 15 K is shown as the solid line, it is fit well by a combination of the three Gaussian peaks (shown as dashed lines) labelled A, B and C. (b) Arrhenius plot of the three Gaussian, with the lines showing their fitting by the Arrhenius dissociation model.

© 2005 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

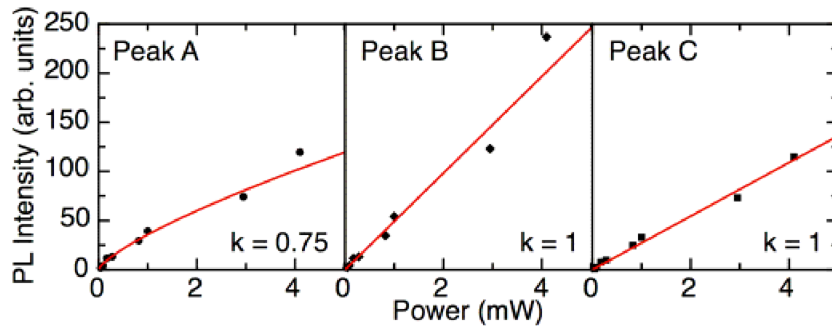

Fig. 2 PL intensity of the three peaks plotted versus pump power, P. The intensity of both Peak B and Peak C has a linear dependence on pump power, while the intensity of Peak A is proportional to $P^{0.75}$.

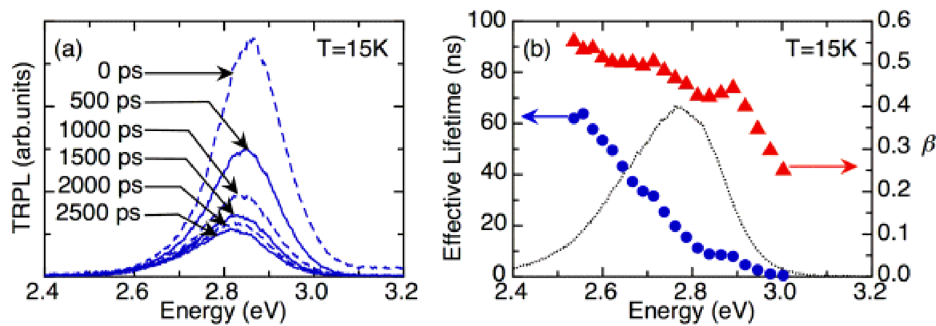

Fig. 3 (a) PL at every 500 ps after pulsed excitation. (b) Plot of the effective lifetime τ and the stretch exponential parameter β versus emission energy. The time-integrated PL is plotted in the background as reference.

ing that these two peaks might be excitonic in nature. Moreover, the existence of two excitonic emission peaks can be the result of In-segregation, and the two peaks may correspond to two different In-phases.

To further identify the excitonic nature, power dependent studies were conducted. The power studies under CW excitation at 15 K shows that these peaks have different pump power dependencies. The relationship between I and pump power, P, can help to determine the nature of the carriers involved in the emission. More specifically, the power law model, $I \propto P^k$, can identify if the emission is excitonic like or if it involves bound states [12]. Figure 2 shows the dependence of I on P of the three peaks: for A the dependence is nonlinear with $k = 0.75$, indicating that the emission involves a bound state; while for B and C, the dependence is linear ($k = 1$), confirming that these peaks are from free exciton emission.

The TRPL time slices at 15 K are shown in Fig. 3(a). Immediately after the pulsed excitation, the PL peaked at 2.86 eV, corresponding to Peak C. As time passes, the emission peak shifted towards the low energy (red shift) and the lifetime of emission from this low energy state was very long lived. This red shift is the result of carrier relaxation. Moreover, the PL decay was non-exponential and is well fitted with the stretch exponential function, $I(t) = I(0)\exp[-(t/\tau)^{\beta}]$, where τ is the effective lifetime and the parameter β is the stretched exponential coefficient. This function, which describes the non-exponential decay of disordered systems, has been very successful in modelling the PL decay of InGaN [13, 14]. This success is due to the fundamental assumption that potential fluctuations exist inside the material, which is the case in many InGaN samples. In this model, the carriers are trapped by the potential minima of the fluctuations and can only move by hoping from one trap to another before recombination. Quantitatively, β falls between 0 and 1, and the smaller the β value the more carriers have to hop before recombination; whereas as β approaches unity the recombination becomes single exponential since no hopping is necessary. Figure 3(b) shows the values of τ and β for different emission energies across the PL. Consistent with the observed red shift, τ decreased from low energy to high energy. It is interesting to note

© 2005 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

that there are two small plateaus of τ around 2.70 eV and 2.85 eV corresponding to the peak energies of Peak B and Peak C. The value of β also decreases with emission energy. Moreover, this decrease is more rapid from 2.9 eV upwards, indicating that carrier hopping is much more dominant than recombination above 2.9 eV. However, on the low energy side, the carriers are more localized resulting in less hopping probability and a corresponding higher value of β.

Finally, in the spatially resolved PL experiment, we selectively excite the material with a lower bandgap. The excitation energy of the pump was at 2.71 eV, and the PL spectrum is shown in Fig 4(a). The emission feature at 2.65 clearly corresponds to Peak B, and we attribute the feature at 2.46 eV to deep level emission. An image of the deep level PL was taken using a low pass filter with cut-off energy at 2.43 eV, and is shown in Fig. 4(b), revealing clustering of the deep levels.

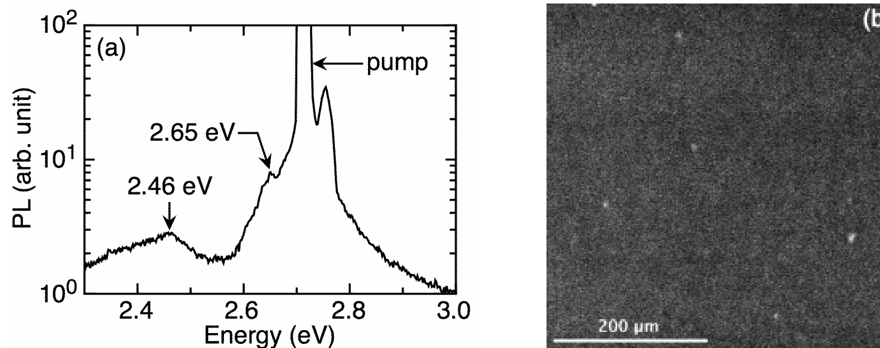

Fig. 4 (a) PL taken under the confocal microscope without the long pass filter. (b) Confocal microscope image of the PL: a long pass filter with cut-off at 510 nm (2.43 eV) was used when obtaining this image.

4 Conclusion We have studied the PL and TRPL characteristics of a Si:InGaN thin film sample grown to facilitate In-Segregation. This sample showed a broad emission composed of three Gaussian peaks. From the results of the PL temperature and power studies, we have tentatively attributed the lowest energy peak to deep level emission, while the two high-energy peaks are excitonic in nature. The two excitonic peaks are more than 100meV apart. Low temperature PL decay was well fitted with a stretched exponential, consistent with disorder in the sample. Finally, using a confocal microscope system, we have found evidence of clustered deep level emission.

Acknowledgements This research was in part supported by ANC's NSF CAREER Grant #9733720, ONR YIP Grant # N00014-00-1-0508, and Doolittle's ONR Grant #N00014-99-1-0419.

References

[1] S. Nakamura and G. Fasol, The blue laser diode: GaN based light emitters and lasers (Springer, Berlin, New York, 1997).
[2] Y. Narukawa et al., J. Cryst. Growth **190**, 606 (1998).
[3] C. A. Tran et al., J. Cryst. Growth **195**, 397 (1998).
[4] K. P. O'Donnell, R. W. Martin, and P. G. Middleton, Phys. Rev. Lett. **82**, 237 (1999).
[5] A. A. Yamaguchi, Y. Mochizuki and M. Mizuta, Jpn. J. Appl. Phys. Part 1 **39**, 2402 (2000).
[6] H. Jiang et al., IEEE J. Quantum Electron. **35**, 1483 (1999).
[7] A. N. Cartwright et al., MRS Internet J. Nitride Semicond. Res. (1999).
[8] L. H. Peng, C. W. Chuang, and L. H. Lou, Appl. Phys. Lett. **74**, 795 (1999).
[9] F. Chen et al., Solid State Commun. **125**, 617 (2003).
[10] T. Onuma et al., Jpn. J. Appl. Phys. Part 1 **42**, 7276 (2003).
[11] M. Leroux et al., J. Appl. Phys. **86**, 3721 (1999).
[12] T. Schmidt, K. Lischka, and W. Zulehner, Phys. Rev. B **45**, 8989 (1992).
[13] M. Pophristic et al., Appl. Phys. Lett. **73**, 3550 (1998).
[14] M. Pophristic et al., MRS Internet J. Nitride Semicond. Res. (2000).

© 2005 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim