Time-Resolved Photoluminescence Studies of Indium-Rich InGaN Alloys *

CHEN Guang-De(陈光德)[1], ZHU You-Zhang(竹有章)[1**], YAN Guo-Jun(颜国君)[1], YUAN Jin-She(苑进社)[1], K. H. Kim[2], J. Y. Lin[2], H. X. Jiang[2]

[1]*Department of Applied Physics, Xi'an Jiaotong University, Xi'an 710049*
[2]*Department of Physics, Kansas State University, Manhattan, Kansas 66506-2601, USA*

(Received 23 September 2004)

Time-resolved photoluminescence (PL) spectroscopy has be used to investigate indium-rich InGaN alloys grown on sapphire substrates by metal organic chemical vapor deposition. Photoluminescence measurement indicates two dominant emission lines originating from phase-separated high- and low-indium-content regions. Temperature and excitation intensity dependence of the two main emission lines in these InGaN alloys have been measured. Temperature and energy dependence of PL decay lifetime show clearly different decay behaviour for the two main lines. Our results show that photo-excited carriers are deeply localized in the high indium regions while photo-excited carriers can be transferred within the low-indium-content regions as well as to high-content regions.

PACS: 78. 47. +p, 78. 55. Cr

In recent years, Ⅲ-nitride semiconductors have attracted much attention due to their applications in electronic and optoelectronic devices such as high brightness blue and green light emitting diodes (LEDs) and laser diodes (LDs).[1−3] InGaN has been used as active layers for most nitride LEDs.[1,3] Although InGaN/GaN quantum-well (QW) LEDs have been commercialized, the growth and understanding of InGaN alloys with high In content is still in the early stages.[4−7] It is important to understand the fundamental properties of the optical process and emission mechanism in InGaN alloys, especially for In-rich InGaN alloys for further improvement of these devices as well as for more devices in optoelectronic applications.

Phase segregation and separation in InGaN alloys is an interesting and important phenomenon, which has attracted great attention.[8,9] In this Letter, we present the results of time-resolved photoluminescence (TRPL) studies of In-rich $In_xGa_{1-x}N$ alloys. Two emission lines at 464 nm (2.672 eV) and 678 nm (1.827 eV) were measured in the samples. Temperature and excitation intensity dependences of these two main emission lines of the InGaN alloys have been measured. Temperature and energy dependence of PL decay lifetime for both the emission lines show different characteristics. Our results confirm the phase separation in the In-rich InGaN alloys. More importantly, we find that, due to separation, PL spectra as well as emission properties depend strongly on temperature for this material, and photo-excited carriers in this material can also be transferred within the low indium regions as well as to high indium regions.

The $In_xGa_{1-x}N$ samples were grown by metal organic chemical vapour deposition (MOCVD) on sapphire substrates with a low temperature buffer. The growth temperature and pressure were 800°C and 80 Torr, respectively. Trimethylgallium (TMG), trimethylindium and ammonia were used as the source precursors for Ga, In, and N, respectively. The thickness of InGaN epilayers is about 0.1 μm, which is grown on the top of 1 μm GaN. The average In composition was roughly 32%. Photoluminescence (PL) spectra were measured with a picosecond time-resolved PL system.[10]

Figure 1 shows the cw PL emission spectra of the InGaN sample measured at three representative temperatures. The two main emission lines at 464 nm (2.672 eV) and 678 nm (1.827 eV) were clearly observed and several features can be obtained from their intensity temperature dependence. (1) At 300 K, emission line at 678 nm is dominant with its intensity about 4 times larger than that at 464 nm. (2) While temperature decreases, the intensity of emission line at 464 nm increases, and so does that of emission line at 678 nm. When temperature decreases to 90 K, the intensity of emission line at 678 nm is still comparable, but at 10 K the intensity of emission line at 464 nm is dominant. We suggest that the two lines originate from two phase segregation or different size quantum dots (QDs), where the blue line is due to small size QDs or low-indium region and the red line is from larger-sized QDs or high-indium-content region. This is in agreement with the results of other research groups.[4,11] The wavelength of the emission peak for both the lines shows almost no variation with the increase of the temperature, compared to the well known shift of the band edge and also to the S-shape variation in InGaN QWs, something which we cannot suitably interpret at present, although the same

* Supported by the National Natural Science Foundation of China under Grant No 10474078.
** Email: yzh_zhu@sina.com

results have been reported before.

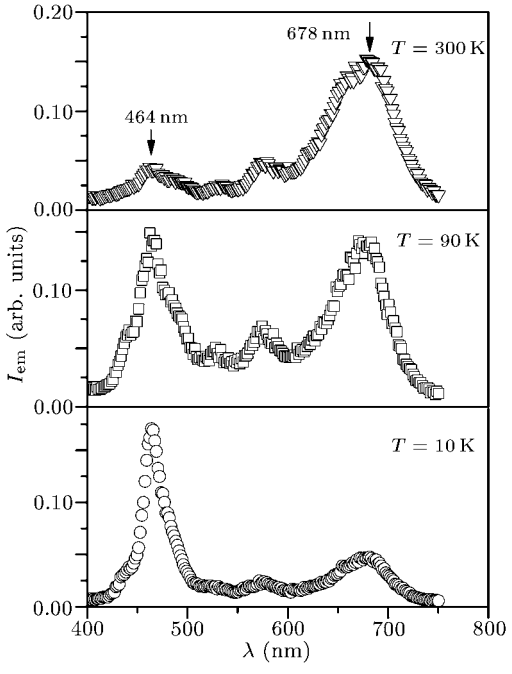

Fig. 1. Continuous-wave PL emission spectra of the $In_xGa_{1-x}N$ sample measured at three typical temperatures.

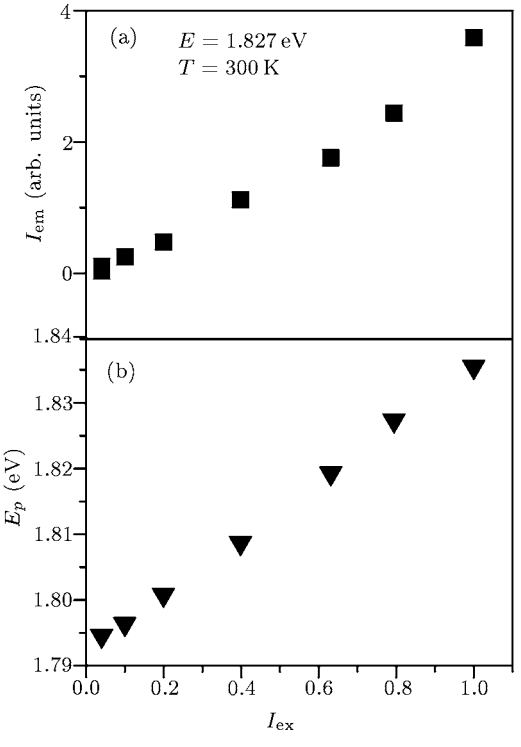

Fig. 2. Excitation intensity dependence of PL emission intensity and peak positions of red line.

Figure 2 shows the excitation intensity dependence of the peak position and the emission intensity for red peak (678 nm) at temperature 300 K. Both the peak position and emission intensity increase linearly with the increase of the excitation intensity. The fact that the energy positions increase linearly with the increase of the excitation intensity can be interpreted as weak Coulomb screening of the quantum confined Stark effect induced by the weak piezoelectric field due to the thickness of our sample in thickness 0.1 μm.

Recombination dynamics of the PL emission of the sample has also been studied. Figure 3 shows the temporal responses of the 464 nm and 678 nm emission lines at 10 K. As shown in Fig. 3, the PL decay temporal responses at the peak 464 nm can be fitted quite well by a single exponential function, but that at the line 678 nm must be fitted by two exponential functions. This shows that the recombination channels of the two lines are different.

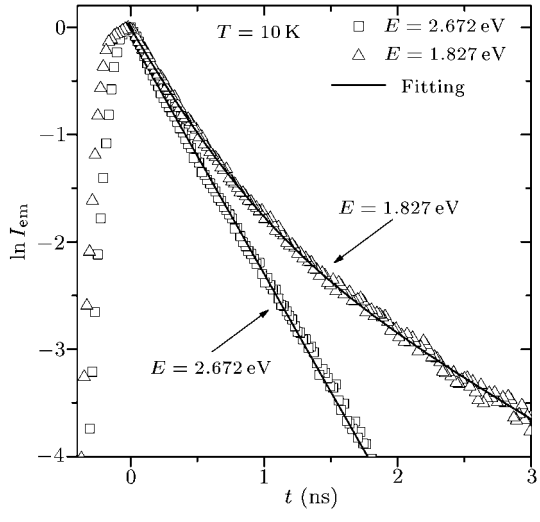

Fig. 3. Temporal responses of the 464 and 678 nm emission lines measured at 10 K.

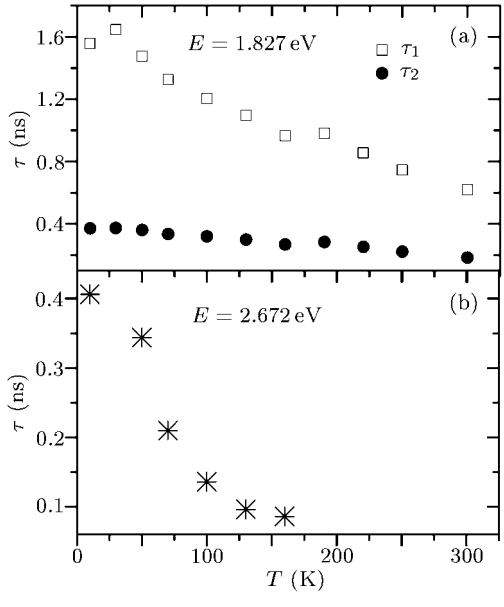

Fig. 4. Temperature dependence of PL decay lifetime measured at spectra peak position of $E = 1.827$ eV and $E = 2.672$ eV.

The temperature dependence of the PL decay time measured both at 464 nm and at 678 nm are plotted in Fig. 4. We have measured the emission energy dependence of the PL decay time for both the peaks of the sample at the temperature 10 K as shown in Fig. 5, and the two representative emission spectra are shown in Fig. 4. The emission energy dependence of the PL decay is a characteristic of a distribution of localized excitons.[10,12,13] Decay data of the blue emission line in Fig. 4(a) are fitted with the least squares method by using the function

$$\tau(E) = \tau_0 + \tau/\{1 + \exp[\alpha(E - E_m)]\}. \qquad (1)$$

The fitting parameters are $\tau_0 = 0.0249$ ns, $\tau = 0.474$ ns, $E_m = 2.713$ eV, and $\alpha = 0.269$ (1/eV), respectively. The value of E_m is approximately 40 meV above the 10 K PL emission peak, which represents the behaviour of strongly localized excitons.[14]

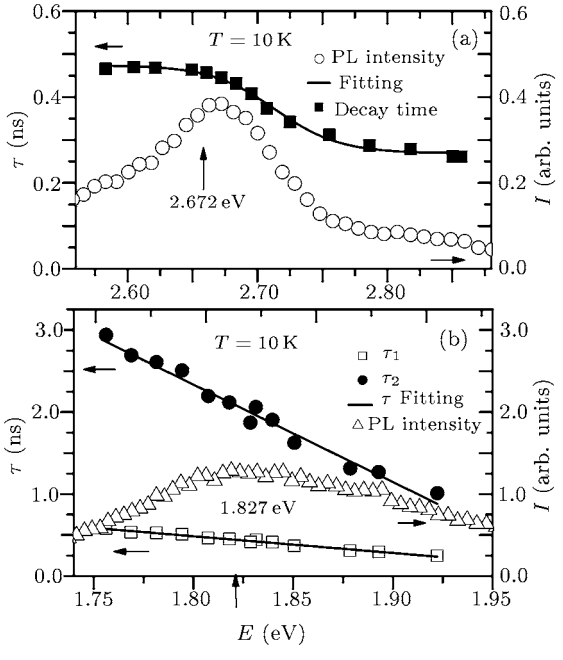

Fig. 5. Emission energy dependence of PL decay lifetime of two main emission lines measured at 10 K.

Decay data of the red emission line in Fig. 4(b) are fit with the least squares method by using of the formula

$$\tau(E) = \tau_0 - \alpha E. \qquad (2)$$

The fitting parameters are $\tau_{10} = 4.153$ ns, $\alpha_1 = 2.037$ (1/eV); $\tau_{20} = 23.76$ ns, and $\alpha_2 = 11.90$ (1/eV). The behaviour of PL decay lifetime of the red line is different from that of the blue line, due to different recombination channels. We can conclude that photo-excited carriers can transfer within the low indium regions as well as to the high-indium-content regions.[14]

In conclusion, indium-rich InGaN alloys samples grown on sapphire substrates by MOCVD has been studied by TRPL. Two dominant emission lines originating from phase separation have been observed and investigated. The PL decay lifetime behaviour shows that the photo-excited carriers in these materials can be transferred within the low indium regions as well as to high-content regions. This carrier-transportation mechanism is important to improve the brightness and to enhance the luminescence efficiency for In-rich optoelectronic devices.[15]

References

[1] Nakamura S, Pearton S and Fasol G 2000 *The Blue Laser Diode* (New York: Springer)
[2] O'Donnell K P, Martin R W and Middleton P G 1999 *Phys. Rev. Lett.* **82** 237
[3] Zhou J and Zhang G Y 2002 *Chin. Phys. Lett.* **19** 707
[4] Li Z H, Yang Z J, Qin Z X, Tong Y Z, Yu T J, Lu S, Yang H and Zhang G Y 2003 *Chin. Phys. Lett.* **20** 1350
[5] Chen H J, Feenstra R M, Northrup J E, Aywietz T and Neugebauer J 2000 *Phys. Rev. Lett.* **85** 1902
[6] Mukai T, Yamada M and Nakamura S 1999 *Jpn. J. Appl. Phys.* **38** 3976
[7] Davidson J A, Dawson P, Wang T, Sugahara T, Orton J W and Sakai S 2000 *Semiconduct. Sci. Technol.* **15** 497
[8] Osamura K, Naka S and Murakami Y 1975 *J. Appl. Phys.* **46** 3432
[9] Ho I H and Stringfellow G B 1996 *Appl. Phys. Lett.* **69** 2701
[10] Smith M, Chen G D, Lin J Y and Jiang H X 1996 *Appl. Phys. Lett.* **69** 2837
[11] Husberg O, Khartchenko A, As D J, Vogelsang H, Frey T, Schikora D, Lischka K, Noriega O C, Tabata A and Leite J R 2001 *Appl. Phys. Lett.* **79** 1243
[12] Li W, Bergman P, Monemar B, Amano H and Akasaki I 1997 *J. Appl. Phys.* **81** 1005
[13] Oueslati M, Benoit C and Zouaghi M 1988 *Phys. Rev.* B **37** 3037
[14] Mair R A, Lin J Y, Jiang H X, Jones E D, Alleman A A and Kurtz S R 2000 *Appl. Phys. Lett.* **76** 188
[15] Chichibu S, Sota T, Wada K and Nakamura S 1998 *J. Vac. Sci. Technol.* B **16** 2204