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Growth Condition Dependence of the Photoluminescence Properties of $In_xGa_{1-x}N/In_yGa_{1-y}N$ Multiple Quantum Wells Grown by MOCVD

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1. Introduction

In recently developed high-brightness blue light-emitting diodes and lasers, InGaN is extensively used as the active layer [1]. However the growth of InGaN and its heterostructures is challenging, and the characteristics of these important materials are little understood so samples of good optical quality are sought for research in to their fundamental properties. Only in the last couple of years have detailed and successful growth studies begun to be reported, and there remain few reports involving optical characterization. By atmospheric pressure (AP) MOCVD we have already achieved InGaN heterostructures of high optical quality [2]. Here we present a systematic investigation of the effect of growth conditions on the photoluminescence properties of $In_xGa_{1-x}N/In_yGa_{1-y}N$ multiple quantum wells.

2. Samples

Samples containing three $In_xGa_{1-x}N/In_yGa_{1-y}N$ quantum wells were grown in a two-flow AP MOCVD system. On a sapphire substrate a 30 nm GaN nucleation layer was grown at 500°C then a 2 - 3 μm GaN layer at 1075°C. InGaN barrier (3 nm) then well (5 nm) materials were each deposited three times before a final barrier layer. Trimethylgallium (TMG), trimethylindium (TMI) and ammonia (NH_3) were used as sources of Ga, In and N respectively, with a mixed H_2/N_2 carrier gas. In later samples silicon doping was provided by SiH_4. Table I gives details of the growth parameters for the InGaN quantum well region for several series of samples. Using these series in which just one growth parameter was varied at a time we have carefully investigated the effects of different factors. An atomic force microscope and a Nomarski optical microscope were used to check surface morphology.

3. Results

Photoluminescence was measured at room temperature (RT) and 77 K. The excitation source was a He-Cd laser (325 nm). Collected light was analyzed using a 0.3 m monochromator and a CCD camera. The high quality of the samples was evidenced by their narrow PL line widths, generally 7 - 13 nm at room temperature. The ratio of band edge to deep level emission peak intensity ranged from approximately 10 to as high as 1000 at RT and more at 77 K. At 77 K the peak luminescence intensity of almost all samples was very similar; the intensity at RT is a better indicator of sample quality since at this temperature non-radiative processes will dominate in poorer samples.

It is now well established that for InGaN lower growth temperatures give a greater incorporation of indium [1], and we have seen this in our samples. However higher temperatures give better crystal morphology, so it is preferable to increase the indium content by optimizing other factors. Raising the TMI flow rate increases indium content as shown in Fig. 1, where the wavelength of the PL peak is plotted against the TMI flow rate for the well material. (Larger wavelength indicates greater indium content.) The quantities and relative proportions of the carrier gases used are also important. We have investigated the influence of the ammonia and hydrogen flow rates and the total gas flow rate.

Increasing the flow rate of NH_3 was found to significantly increase both the intensity and wavelength of the PL. This is shown in Fig. 2. Results for H_2 are shown in Fig. 3 and indicate that a moderate flow of this gas is best. One group has reported an increase in the ratio of band edge to deep level emission with increased NH_3 flow [3]. However others have found the indium content of AP MOCVD grown InGaN films to be little affected by the NH_3 flow rate but increased by lowering the H_2 flow rate [4,5]. The reason for this difference in results is as yet unclear. Fig. 4 shows that decreasing the total gas flow rate also gives greater PL intensity and wavelength.

When all flow rates were halved, both wavelength and intensity were severely reduced (note the low intensities of series E, Table I and Fig. 2). However when the growth rate was reduced, by quartering the flow rates only of TMG and SiH_4, the resulting PL had shorter wavelength but significantly higher intensity: the RT PL of all such samples was consistently as strong as in the best previous sample and the intensity at 77 K was greater by a factor of about three. Fig. 5 shows the PL spectrum of one of these samples.

4. Conclusions

$In_xGa_{1-x}N/In_yGa_{1-y}N$ triple quantum wells of high optical quality have been grown by atmospheric pressure MOCVD and characterized by photoluminescence. By systematic investigation of different growth parameters we conclude that optimum growth conditions include high ammonium flow rate, moderate hydrogen flow rate, low total gas flow rate and low growth rate. Room temperature PL line widths as narrow as 7 nm have been achieved.

References

1) For a review, S. Nakamura and G. Fasol: *The Blue Laser Diode* (Springer-Verlag, Berlin, 1997).
2) T. Someya, K. Tachibana, M. Nishioka and Y. Arakawa: to be published in Proc. 17th Electronic Materials Symposium.
3) W. Van der Stricht, I. Moerman, P. Deemster, L. Considine, E. J. Thrush and J. A. Crawley: Internet J. Nitride Semicond. Res. **2** (1997) 16.
4) S. Keller, B. P. Keller, D. Kapolnek, U. K. Mishra, S. P. DenDaars, I. K. Shmagin, R. M. Kolbas, S. Krishnankutty: J. Crystal Growth **170** (1997) 349.
5) E. L. Piner, M. K. Behbehani, N. A. El-Masry, F. G. McIntosh, J. C. Roberts, K. S. Boutros and S. M. Bedair: Appl. Phys. Lett. **70** (1997) 461.

Series	Growth Temperature (°C)	Gas Flow Rates TMG (μmol/min)	TMI/barrier (μmol/min)	TMI /well (μmol/min)	NH_3 (l/min)	H_2 (cc/min)	SiH_4 (cc/min)	Total (l/min)
A	770	44	5.33	*	10	200	0	22.5
B	765	44	5.33	*	10	200	10	22.5
C	770	44	5.33	*	10	200	10	22.5
D	775	44	5.33	*	10	200	10	22.5
E	770	22	2.67	20	*	150	0	22.5
F	750	44	5.33	40	*	300	0	18
G	770	44	5.33	40	10	*	0	22.5
H	770	44	5.33	40	10	300	0	*
Sample J	780	5.5	5.33	40	10	200	1.3	22.5

Table I Growth parameters for the $In_xGa_{1-x}N/In_yGa_{1-y}N$ quantum wells in several series of samples. * marks the parameter varied in each series.

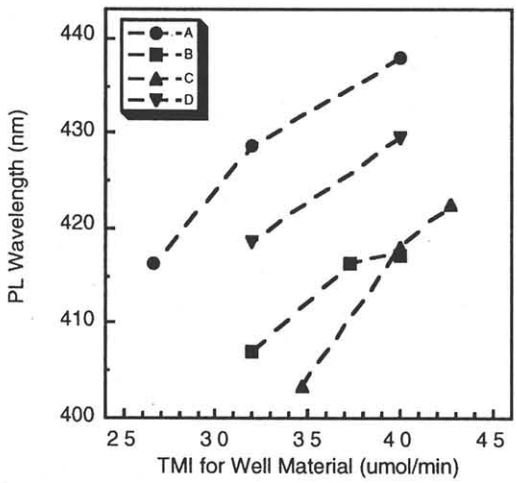

Fig. 1 Influence of TMI flow rate for well material on peak wavelength of RT PL in samples of series A - D. Lines are a guide to the eye.

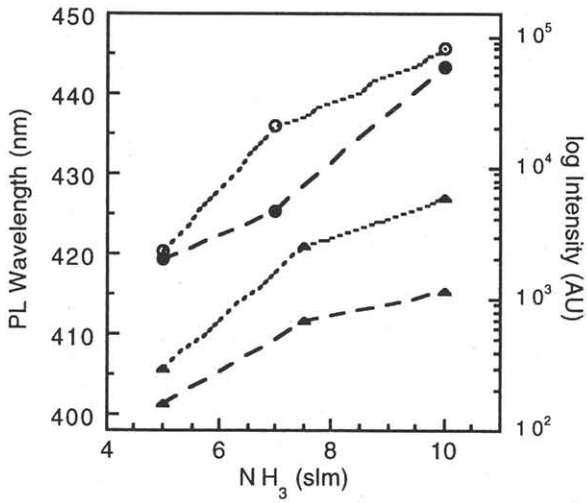

Fig. 2 Influence of ammonia flow rate on peak wavelength (solid symbols) and intensity (open symbols) of RT PL in samples of series E (triangles) and F (circles). Lines are a guide to the eye.

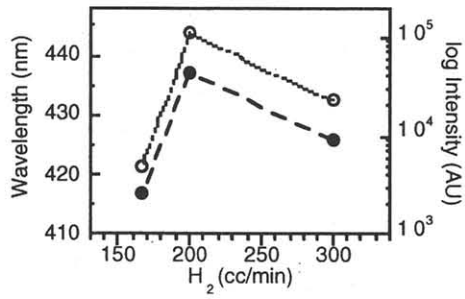

Fig. 3 Influence of hydrogen flow rate on peak wavelength (solid symbols) and intensity (open symbols) of RT PL in sample series G. Lines are a guide to the eye.

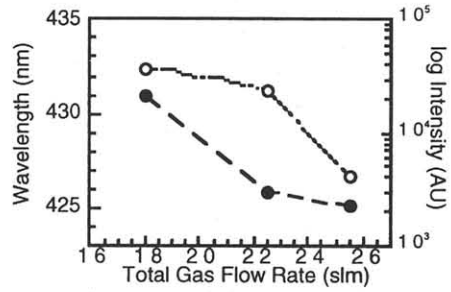

Fig. 4 Influence of total gas flow rate on peak wavelength (solid symbols) and intensity (open symbols) of RT PL in sample series H. Lines are a guide to the eye.

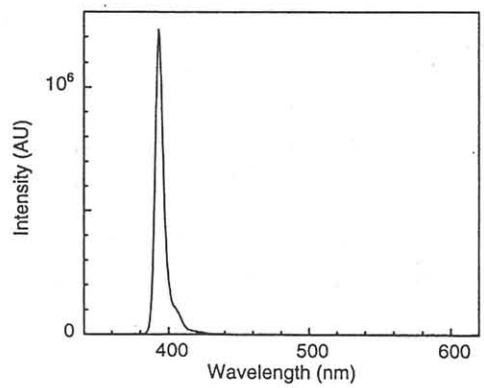

Fig 5. Photoluminescence spectrum of sample J at 77K. The FWHM of the peak is 6 nm.