Effects of InGaN barriers with low indium content on internal quantum efficiency of blue InGaN multiple quantum wells*

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Blue $In_{0.2}Ga_{0.8}N$ multiple quantum wells (MQWs) with $In_xGa_{1-x}N$ ($x = 0.01 - 0.04$) barriers are grown by metal organic vapour phase epitaxy. The internal quantum efficiencies (IQEs) of these MQWs are studied in a way of temperature-dependent photoluminescence spectra. Furthermore, a 2-channel Arrhenius model is used to analyse the nonradiative recombination centres (NRCs). It is found that by adopting the InGaN barrier beneath the lowest well, it is possible to reduce the strain hence the NRCs in InGaN MQWs. By optimizing the thickness and the indium content of the InGaN barriers, the IQEs of InGaN/InGaN MQWs can be increased by about 2.5 times compared with conventional InGaN/GaN MQWs. On the other hand, the incorporation of indium atoms into the intermediate barriers between adjacent wells does not improve IQE obviously. In addition, the indium content of the intermediate barriers should match with that of the lowest barrier to avoid relaxation.

Keywords: metal organic vapour phase epitaxy, quantum wells, nitrides, light emitting diodes
PACC: 6855, 7280E, 7865K, 8115

1. Introduction

Recently, solid state lighting utilizing InGaN light emitting diodes (LEDs) have demonstrated great advantages in energy conservation.[1−3] However, their internal quantum efficiencies (IQEs) are still a bottleneck for further improvement in lumen efficiency. The InGaN/GaN multiple quantum wells (MQWs), widely used as the active layers in LEDs, often show poor crystalline quality and strong piezoelectric field.[4−9] The poor crystalline quality of InGaN/GaN MQWs leading to multiple nonradiative recombination centres (NRCs) is mostly induced by hetero-epitaxy on sapphire substrates,[4] low growth temperature,[5] and large lattice mismatch between InN and GaN.[6] The strain-induced piezoelectric field in InGaN QW can result in the spatial separation of electron and hole wave functions or quantum-confined Stark effect (QCSE),[7−9] which leads the wavelength to have a redshift and radiative recombination rate to be reduced.

In the previous studies,[10−12] the replacement of GaN barriers with InGaN barriers was demonstrated to be an efficient technique to improve the IQEs of MQWs. Generally, the barrier beneath the lowest quantum well, which is referred to as LB in this paper, is different from the intermediate barrier (IB) between adjacent wells. Because the LB is adjacent to the growth interruption interface between GaN bulk layer and MQW grown at a lower temperature, its thickness is usually large enough to eliminate interface effects. Akasaka *et al.*[10] obtained a near-ultraviolet InGaN MQW structure, wherein the LB and IBs were 50 and 5-nm-thick $In_{0.04}Ga_{0.96}N$ layers, respectively. Its IQE was found to be as high as 71% at room temperature. Hence it was estimated that the NRCs were well eliminated by the incorporation of indium atoms into the LB. Other researchers[13−15] also tried to grow InGaN/GaN MQWs with GaN IBs, but the LB was entirely or partially replaced by InGaN layer. However, the InGaN LB changes the strain in InGaN MQWs,

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leading to changes in optical characteristics.[13−15] Recently, Morkoç *et al.*[16−18] found that reducing the IB height was favourable for efficient hole transport through the active region, which consequently reduced the efficiency droop effect. That means that the incorporation of indium into IBs will facilitate hole injection, while it will also influence the IQEs of the MQWs. However, to the knowledge of the authors, there were few publications on the effects of the thickness and the indium content of InGaN barriers on the IQEs of InGaN MQWs.

In the present work, blue InGaN MQWs with various LBs and IBs of are grown and investigated by temperature-dependent photoluminescence (PL) measurements. After fitting with a 2-channel Arrhenius model, the effects of InGaN barriers on IQE and NRCs of the MQWs are analysed.

2. Experiment

The samples are grown on c-plane sapphire substrates by the Aixtron 2000HT metal organic vapour phase epitaxy (MOVPE) system. Trimethylgallium (TMGa), triethylgallium (TEGa), trimethylindium (TMIn), and NH_3 are used as the precursors of Ga, In, and N, respectively. First, the substrates are cleaned in hydrogen ambience at 1080 °C for 10 min. Next, a 30–nm-thick GaN buffer layer is deposited at 530 °C followed by a 4-μm-thick GaN bulk layer grown at 1050 °C. Then, the carrier gas is changed from hydrogen into nitrogen and the growth temperature is decreased to 740 °C for the growth of InGaN MQWs. An InGaN LB and a 5-period $In_{0.2}Ga_{0.8}N$ MQWs structure are then grown at the same temperature. The thicknesses of the well and IB are 2 and 22 nm, respectively. Finally, a 5-10 nm GaN capping layer is grown on the top of the wafer. The indium content is controlled by the flow rate of TMIn and determined by fitting the high-resolution x-ray diffraction rocking curves. Ten samples with different values of thickness and indium content of InGaN barriers are grown, with their main parameters listed in Table 1.

Table 1. Barrier parameters, IQEs, and fitting parameters of the samples.

sample No.	thickness of the LB/nm	indium content in the LB	indium content in the IBs	IQE	N_1	E_1/meV	N_2	E_2/meV
A	60	0	0	21.3%	10.9	28.6	0.43	3.6
B	30	0.01	0.01	33.5%	6.1	30.3	0.18	2.9
C	60	0.01	0.01	49.5%	3.5	31.2	0.06	1.7
D	90	0.01	0.01	50.0%	3.4	32.3	0.03	1.9
E	60	0.02	0.02	41.0%	6.9	42.0	0.05	1.7
F	30	0.04	0.04	24.5%	12.5	38.9	0.17	5.2
G	60	0.04	0.04	37.8%	11.0	51.6	0.04	1.9
H	90	0.04	0.04	23.5%	19.4	53.4	0.36	7.9
J	60	0.01	0	49.5%	3.9	33.5	0.05	1.9
K	60	0.01	0.04	19.8%	125.8	90.5	0.84	8.4

After growth, temperature dependent PL spectra of all the samples are measured from 10 K to room temperature (RT), with a 325 nm He–Cd laser serving as an excitation source.

3. Results and discussion

The PL wavelengths at RT of Samples A–H are shown in Fig. 1, and the PL spectra at RT of samples A and C are exemplified in the inset. Compared with the PL peak of sample A, the PL peaks of samples with InGaN barriers each exhibit a blueshift, which is attributed to the reduction in strain hence the QCSEs in InGaN wells. As shown in Fig. 1, this blueshift in PL wavelength increases with the increase of thickness of InGaN LB or indium content in InGaN barriers. According to the previous work,[10,11,13] the strain relief function can be attributed to the InGaN LB. However, it is hard to distinguish the contribution from the IBs, which can also act as strain relief layers theoretically. The InGaN IBs will be discussed later.

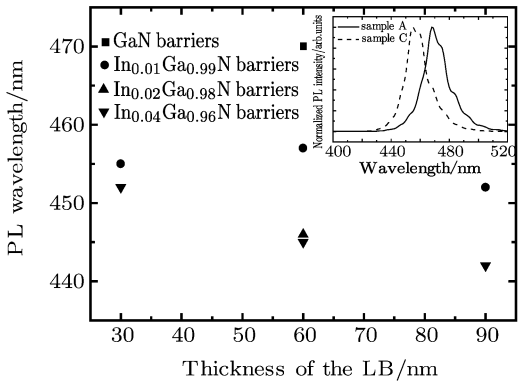


Fig. 1. PL wavelengths for samples A–H at RT. The inset shows normalized PL spectra of samples A and C at RT.

As a common practice, the IQEs of all the samples are each defined as the ratio of integral PL intensity at RT to that at 10 K,[10,11] and are listed in Table 1. The IQE values show they are dependent on both the thickness and the indium content of the InGaN barriers. In order to explore IQE variation, a 2-channel Arrhenius model is used to fit the temperature dependent integral PL intensity.[19,20] Assuming that the thermal activation of NRC is the main mechanism for PL quenching at high temperatures and that there exist two types of NRCs in MQWs, the normalized PL intensity can be expressed as

$$I(T) = \frac{1}{1 + N_1 \exp\left(-\dfrac{E_1}{k_{\mathrm{B}}T}\right) + N_2 \exp\left(-\dfrac{E_2}{k_{\mathrm{B}}T}\right)},$$

where N_1 and N_2 represent the rate constants related to NRC densities, E_1 and E_2 are the activation energies of the first- and second-type NRCs, k_{B} is the Boltzmann constant, and T is the temperature. Fitting parameters for different samples are also listed in Table 1, and the Arrhenius plots of samples A and C are shown in Fig. 2 as examples. The exact origins of these two types of NRCs are still unclear. Generally, NRCs with activation energy between 30 and 60 meV are considered to be related to dislocations in MQWs, while those with activation energy below 10 meV are attributed to carrier-escaping channels located in MQW interfaces.[19,20] By comparing N_1 and N_2 of our samples, it can be concluded that the dominant factor influencing the IQE at RT is the first-type NRC. Thus we focus on the dependence of the first-type NRC on InGaN barriers in this article.

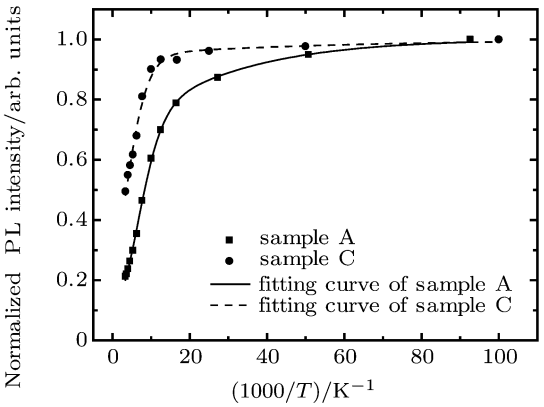


Fig. 2. Arrhenius plots of normalized PL intensity versus temperature of samples A and C.

According to Table 1, samples B–E exhibit simultaneously high E_1 and low N_1 compared with sample A, which certainly results in higher IQE. But samples F–H, all of which have higher E_1 and N_1, also exhibit higher IQEs. These data show that the strain is critical to the density and the activation energy of the NRC, whose formation is related to the strain relaxation during growth.[6,21] In samples B–E, the strains in MQWs are reduced by the InGaN LB, so the density of NRCs also decreases. As the indium content is low, the accumulated stresses in InGaN barriers can be neglected. However, in samples F–H, the accumulated stresses $In_{0.04}Ga_{0.96}N$ barriers will produce new NRCs even though the strains in the MQWs are reduced. It should be noticed that samples C and D have the similar IQEs, which means a 60 nm InGaN LB is thick enough to reduce the strains in MQWs in our samples. On the other hand, the activation energy of the NRC seems to depend on the strain of MQW. In samples with shorter PL wavelengths, the activation energy becomes higher. In the previous work, the NRCs were supposed to be surrounded by potential barriers and carriers confined at low potential locations,[22,23] so the activation energy was considered to be related to the potential fluctuation.[19] In the present case, the potential fluctuation is believed to be dependent on the strain in the InGaN QW. However, further study is desired for more explicit explanation. Nevertheless, the IQE is achieved to be 50% for sample C, which is about 2.5 times higher than that of Sample A.

To confirm the effects of InGaN IBs, wavelengths and IQE of samples A, C, J, G, and K are compared in Fig. 3. Without InGaN IBs, samples J and C have similar optical characteristics, which implies that the

LB has a predominant contribution in strain relief. Meanwhile, it is believed that the elimination of NRCs in our samples is mainly due to the strain reduction rather than the incorporation of indium atoms into GaN,[10] for sample C shows higher IQE than sample J if the later mechanism dominates. In sample K, wherein the LBs and the IBs are different in indium content, the IQE is poor. It is probably because the strain relief provided by the $In_{0.01}Ga_{0.99}N$ LB is not sufficient in this case, and the MQWs become relaxed, thereby producing a lot of NRCs. It can be concluded that the indium content of the LB should be matched with those of IBs, if we need to reduce the barrier height for certain design targets.[17,18]

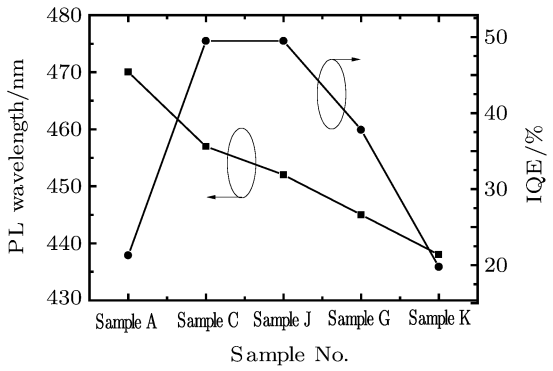


Fig. 3. PL wavelengths and IQE at RT of samples A, C, J, G, and K.

4. Conclusion

It is proved that the InGaN LB has the capacity to reduce the strains in InGaN wells. As a result, the QCSE and the NRCs in InGaN MQWs can be reduced. On the other hand, the accumulated stresses in InGaN barriers will also affect the InGaN MQWs, which should be considered during parameter optimization. The IQE is achieved to be 50% (about 2.5 times higher than that in the conventional InGaN MQWs with GaN barriers) when the thickness and the indium content of the InGaN LB are 60 nm and 0.01, respectively. The incorporation of indium into IBs is found to be inefficient to eliminate NRCs in our samples; but when indium is expected to meet some design targets, its content should be matched with that in the LB to avoid harming IQE.

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