Phys. Status Solidi C **13**, No. 5–6, 209–213 (2016) / **DOI** 10.1002/pssc.201510187

physica status solidi c
www.pss-c.com
current topics in solid state physics

Effect of QW growth temperature on the optical properties of blue and green InGaN/GaN QW structures

Simon Hammersley[*,1], Menno J. Kappers[2], Fabien C.-P. Massabuau[2], Suman-Lata Sahonta[2], Phil Dawson[1], Rachel A. Oliver[2], and Colin J. Humphreys[2]

[1] School of Physics and Astronomy, Photon Science Institute, University of Manchester, M13 9PL, UK
[2] Department of Materials Science and Metallurgy, University of Cambridge, 27 Charles Babbage Road, Cambridge, CB3 0FS, UK

Received 30 September 2015, revised 17 December 2015, accepted 12 January 2016
Published online 3 February 2016

Keywords green gap, InGaN, quantum wells, efficiency, photoluminescence

* Corresponding author: e-mail simon.hammersley@manchester.ac.uk, Phone: +44 161 306 4886, Fax: +44 161 275 1031

In this paper we report on the impact that the quantum well growth temperature has on the internal quantum efficiency and carrier recombination dynamics of two sets of InGaN/GaN multiple quantum well samples, designed to emit at 460 and 530 nm, in which the indium content of the quantum wells within each sample set was maintained. Measurements of the internal quantum efficiency of each sample set showed a systematic variation, with quantum wells grown at a higher temperature exhibiting higher internal quantum efficiency and this variation was preserved at all excitation power densities. By investigating the carrier dynamics at both 10 K and 300 K we were able to attribute this change in internal quantum efficiency to a decrease in the non-radiative recombination rate as the QW growth temperature was increased which we attribute to a decrease in incorporation of the point defects.

1 Introduction Light emitting diodes (LEDs) are now widely used for high efficiency visible light generation, with InGaN quantum wells (QWs) active regions being utilised when emission in the blue and green regions of the visible spectrum is required. However devices which emit green light exhibit much lower external quantum efficiencies (EQEs), of the order of 25 % [1], than their blue emitting counterparts, for which EQEs in excess of 70 % are reported [2]. It is also not possible to produce efficient emission using AlInGaP devices in the green region of the visible spectrum, resulting in a wavelength range where the production of a highly efficient LED is not possible, commonly referred to as the "green gap" [3,4].

In order to achieve emission in the green spectral region using InGaN/GaN based LEDs, the indium composition of the QW needs to be higher than that required for blue emission. This increase in indium composition leads to an increase in the strength of the polarisation induced electric fields across the QWs [5-7]. This, in turn, leads to longer radiative recombination lifetimes for green emitting devices compared to their blue emitting counterparts. It is anticipated that this increase in the radiative recombination lifetime in green emitting samples leads to less efficient competition with non-radiative recombination pathways, resulting in the reduction in EQE for green LEDs. However it has been suggested that this increase in radiative recombination lifetime alone is not sufficient to explain the extent to which the EQE is reduced in green emitting devices [8,9], and that the effects of non-radiative recombination must also be increased.

In order to achieve the indium content required in green-emitting devices the growth temperature of the QW is reduced, from approximately 750 °C for blue emitting devices to approximately 700 °C in green emitting devices. This is to reduce the rate of indium desorption during growth. This lower temperature has been shown to lead to a higher density of structural defects and impurity incorporation [10,11]. Of particular relevance is evidence that a higher density of point defects is introduced when layers of higher indium content are grown at a reduced temperature [13-17], and these point defects have been shown to act as non-radiative recombination centres [18].

In this work we report on how the optical properties of two sets of samples, one set designed to emit at the same wavelength in the blue spectral region and the other at the same wavelength in the green spectral region, are effected by the growth temperature of the QW.

2 Experimental details Six samples, three blue-emitting 10 QW structures and three green-emitting 5 QW structures, were grown by metalorganic vapour phase epitaxy in a 6×2 inch Thomas Swan close-coupled showerhead reactor on GaN pseudo substrates using the two-temperature growth method [19]. The QW and barrier thicknesses were nominally 3 and 7 nm respectively for both sets of samples. Within each set of QWs, the temperature at which the QW was grown was progressively reduced, in order to compensate for this effect the indium precursor flow rate was adjusted in order to maintain a constant indium fraction. X-ray diffraction measurements were used to confirm that the indium compositions and thickness of the QW/barrier repeats were indeed constant for each sample set. A summary of the growth conditions and the results of the X-ray diffraction measurements are shown in Table 1. The X-ray diffraction measurements only provide information as to the average indium content and period of the combined QW and barrier regions due to the presence of gross well width fluctuations introduced by the 2 temperature growth method. Transmission electron microscopy measurements (not shown) did not show any evidence of strain relaxation such as misfit dislocations in any of the samples, indicating that the InGaN QWs are fully strained to the GaN.

For the optical measurements the samples were mounted on the cold finger of a closed cycle helium cryostat with the sample mounted at Brewster angle to the collection optics in order to reduce the effects of Fabry-Pérot interference. Temperature and power dependent photoluminescence (PL) measurements were performed using a continuous wave HeCd laser with a photon energy of 3.815 eV to excite the samples. For the PL decay transients, the frequency tripled output of a mode-locked Ti:sapphire laser, with a final photon energy of 4.881 eV, was used to excite the samples, the PL decay transients were then processed using time correlated single photon counting techniques.

Table 1 Summary of the QW growth conditions and XRD results for the samples discussed in this paper

Sample	QW Growth Temperature (°C)	TMI Flow (μmol/min)	Period (nm)	Indium Content (%)
Blue 1	748	9.7	9.7±0.2	3.2±0.5
Blue 2	730	2.0	9.8±0.2	3.1±0.5
Blue 3	716	1.3	9.7±0.2	3.2±0.5
Green 1	716	9.7	9.9±0.2	5.1±0.5
Green 2	706	4.8	9.9±0.2	5.2±0.5
Green 3	698	3.5	10.0±0.2	5.1±0.5

3 Results and discussion In Fig. 1 is shown the low temperature PL spectra recorded for each of the samples, showing that there is no significant or systematic variation in the PL peak energy or lineshape caused by the variation in QW growth temperature. The green and blue emitting samples have PL emission peaks at 2.4 and 2.75 eV respectively, confirming that the emission wavelength of the QWs within the sample sets were not affected by the changes in the QW growth temperature. The PL spectra of each sample were recorded as a function of excitation power density and temperature, and from this data the 300 K IQE, defined as the ratio of integrated intensity recorded at 300 K compared to that recorded at 10 K, was calculated as a function of excitation power density. The IQEs as a function of power density are shown in Fig. 2. We estimate the range of excitation power densities investigated would be equivalent to a current density range of 1-200 A cm^{-2} in an LED structure. The first thing of note is that for all excitation power densities studied there is a clear trend in the measured 300 K IQEs as a function of QW growth temperature, with lower QW growth temperatures resulting in lower 300 K IQE for both blue and green-emitting samples.

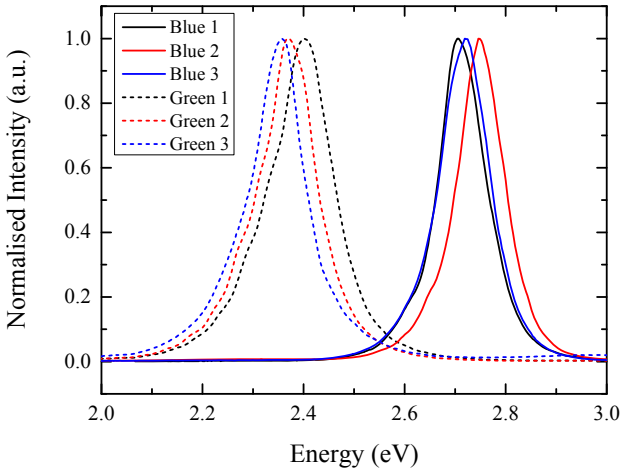

Figure 1 Low temperature (10 K) PL spectra recorded for the blue and green emitting samples

At low excitation power densities all samples showed an increase in 300 K IQE as the excitation power density is increased; this behaviour is widely observed [20-24] and has been attributed to the saturation of the non-radiative recombination pathway as the excitation power density is increased [24]. As the excitation power density is further

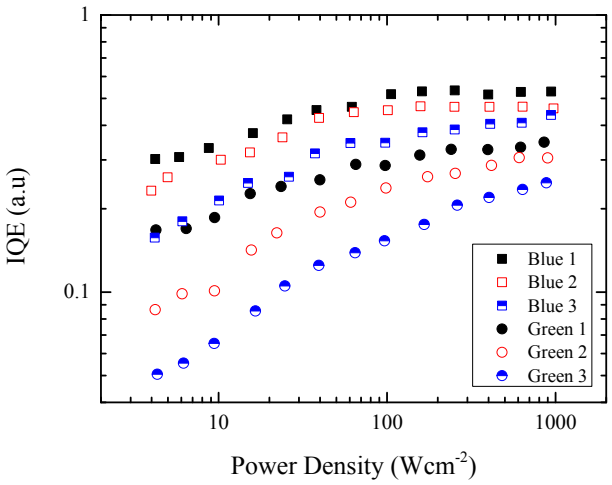

Figure 2 300 K IQE as a function of excitation power density for both the blue and green emitting samples.

increased the rate of increase in IQE slows, in some samples producing a plateau. This behaviour is often observed prior to the onset of the efficiency droop [20-24] which has been reported to occur at a wide range of current densities ranging from 0.2-1000 Acm^{-2} [25,26].

The reduction in IQE for samples grown using lower QW growth temperatures suggests that there is an increase in the non-radiative recombination rate. In order to confirm this hypothesis the 300 K carrier recombination dynamics of the samples must be studied, however at room temperature the recombination of carriers is determined by both the radiative and non-radiative recombination processes within the sample. Therefore in order to conclusively attribute any changes to the carrier recombination dynamics at 300 K to changes in the non-radiative recombination processes, we must ensure that the change in QW growth temperature has not modified the radiative recombination processes. In order to do this the PL decay transients were recorded at 10 K where the recombination is assumed to be purely radiative [27]. The PL decay transients recorded at the PL emission peaks for both the blue and green-emitting samples are shown in Fig. 3(a) and (b), respectively. The time decay measurements were performed at a photoinjected carrier density of 4×10^{11} cm^{-2} QW^{-1} pulse^{-1} and 8×10^{11} cm^{-2} QW^{-1} pulse^{-1} for the blue and green-emitting samples, respectively. This low photoinjected carrier density was chosen in order to avoid any complications caused by efficiency droop related phenomena [20-24].

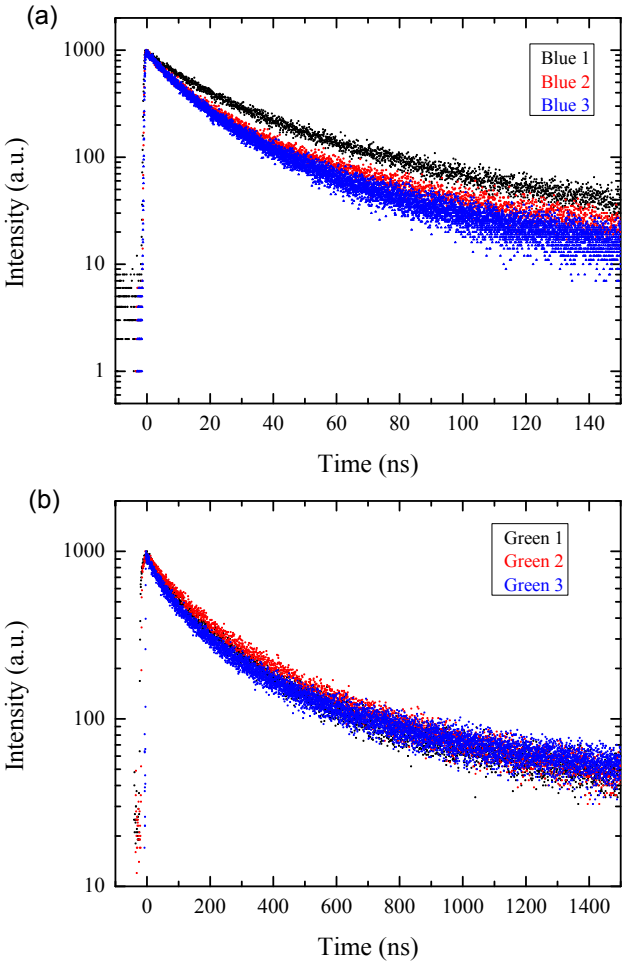

Figure 3 10 K PL decay transients detected at the PL emission peak for the (a) blue and (b) green emitting samples.

All the green-emitting samples have very similar PL decay transients with a measured 1/e PL decay time, defined as the time taken for the PL intensity to fall from its maximum to 1/e of the maximum, of 145 ± 15 ns. For the blue-emitting samples two of the samples Blue 2 and Blue 3 exhibit nearly identical PL decay transients, with 1/e PL decay times of 15 ± 2 ns, however, the observed PL decay transient of sample Blue 1 exhibits a slightly longer 1/e PL decay time of 22 ± 2 ns. The reason for this change in PL decay lifetime is not known, however, this will not affect our conclusions, as a longer PL decay lifetime would lead to a lower IQE if the non-radiative recombination at 300 K is unchanged. From these measurements we therefore conclude that the change in QW growth temperature has not had a significant effect on the radiative recombination rate in either the blue or green-emitting samples.

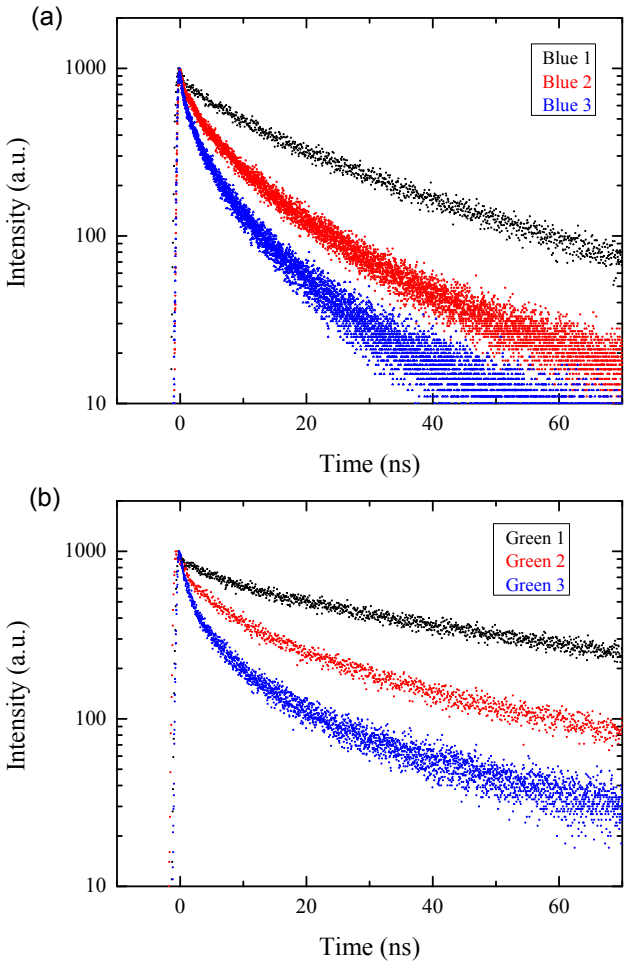

Figure 4 300 K PL decay transients detected at the PL emission peak for the (a) blue and (b) green emitting samples.

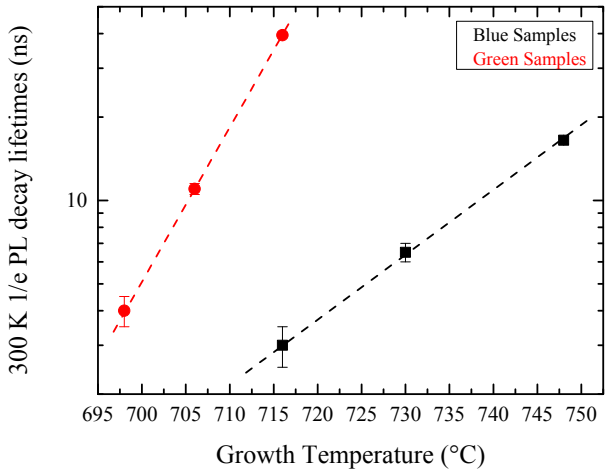

Figure 5 300 K 1/e PL decay times as a function of QW growth temperature.

We now turn our attention to the PL decay transients recorded for the samples at 300 K as shown in Fig. 4. Unlike at 10 K there is now a distinct trend, with the samples grown at lower QW growth temperature, and therefore lower IQE, exhibiting faster PL decay transients for both the blue and green-emitting QW sample sets. The 1/e decay times were found to be 16.5, 6.0, 3.0 ± 0.5 ns for samples Blue 1-Blue 3, respectively, and 39.5, 11.0, 4.0 ± 0.5 ns for samples Green 1-Green 3 (Fig. 5). As at 10 K, where the recombination is assumed to be purely radiative, there was no significant modification of the recombination dynamics as the QW growth temperature was varied, it can therefore be concluded that the variation in PL decay dynamics present at 300 K is a consequence of a change to the non-radiative recombination dynamics between the samples with different QW growth temperatures.

We attribute the observed reduction in the non-radiative recombination rate in samples with lower QW growth temperature to an increase in the density of non-radiative recombination centres incorporated into the samples when lower QW growth temperatures are used [12-15].

4 Summary and conclusions In summary, we have investigated the effect of QW growth temperature on the optical properties of blue and green-emitting QW structures grown at different QW growth temperatures while maintaining the indium composition of the QWs. It was found that at low temperatures (10 K) that modification to the QW growth temperature did not have any significant or systematic effect on the PL emission or the PL decay transients recorded at the PL emission peak. This led us to conclude that the radiative recombination rate within these samples remained unchanged by the QW growth temperature. At 300 K it was found that the samples grown at lower temperature exhibited a lower 300 K PL IQE at all excitation power densities, and a more rapid PL decay transient. This behaviour was found to be systematic, and was attributed to a change in the non-radiative recombination rate. We attributed this reduction in non-radiative recombination lifetime to an increase in the density of point defects incorporated into the crystal due to the reduced QW growth temperature [12-15].

Acknowledgements This work was carried out with the financial support of the United Kingdom Engineering and Physical Sciences Research Council under Grant Nos. EP/I012591/1 and EP/H011676/1.

References

[1] T. Shioda et al., Phys. Status Solidi A **209**, 473-476 (2012).
[2] M. J. Cich et al., Appl. Phys. Lett. **101**, 223509 (2012).
[3] M. Krames et al., J. Display Technol. **3**, 160-175 (2007).
[4] A. Löffler et al., Compd. Semicond. **19**, 32-36 (2013).
[5] Z. Lin et al., Jpn. J. Appl. Phys., Part 1 **54**, 022102 (2015).
[6] K.P. O'Donnell et al,. Phys. Status Solidi RRL **6**, 49-52 (2012).
[7] S.F. Chichibu et al., Appl. Phys. Lett. **69**, 4188-4190 (1996).
[8] T. Langer et al., Phys. Status Solidi C **8**, 2170-2172 (2011).
[9] T. Langer et al., Appl. Phys. Lett. **103**, 022108 (2013).

[10] D. Koleske et al., J. Cryst. Growth **242**, 55-69 (2002).
[11] M.J. Kappers et al., Phys. Status Solidi C **12**, 403-407 (2015).
[12] A. Armstrong et al., J. Electron. Matter. **40**, 369-376 (2011).
[13] E. Gr et al., Appl. Phys. Lett. **99**, 092109 (2011).
[14] A. Armstrong et al., Opt. Express **20**, A812-A821 (2012).
[15] A. Uedono et al., ECS Trans. **61**, 19-30 (2014).
[16] A. Janotti et al,. Phys. Status Solidi A **209**, 65-70 (2012).
[17] T. Obata et al., J. Cryst. Growth **311**, 2772-2775 (2009).
[18] T. Langer et al., Proc. SPIE **8625**, 862522 (2013).
[19] R.A. Oliver et al., Appl. Phys. Lett. **103**, 141114 (2013).
[20] Y.C. Shen et al., Appl. Phys. Lett. **91**, 141101 (2007).
[21] M. –H. Kim et al., Appl. Phys. Lett. **91**, 183507 (2007).
[22] I. Rhozansky et al., Semiconductors **40**, 839-845 (2006).
[23] M.F. Schubert et al., Appl. Phys. Lett. **94**, 081114 (2009).
[24] H. Murotani et al., Phys. Status Solidi B **252**, 940-945 (2015).
[25] A. Hanglieter et al., Proc. SPIE **9363**, 93631R (2015).
[26] J. Xie et al., Appl. Phys. Lett. **93**, 121107 (2008).
[27] C.E. Martinez et al., J. Appl. Phys. **98**, 053509 (2005).