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# High excitation carrier density recombination dynamics of InGaN/GaN quantum well structures: Possible relevance to efficiency droop

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We report on the optical properties of InGaN/GaN quantum well structures measured at 10 K as a function of excitation density. At high excitation power densities we observe a component in the spectra that decays more rapidly than the localised carrier emission observed for low excitation power densities. We attribute this component to recombination involving weakly localised or delocalised carriers. At the high excitation power densities there is a reduction in the recombination internal quantum efficiency, so called efficiency droop. These observations are compatible with the model whereby efficiency droop is explained in terms of the non radiative loss of delocalised carriers. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4781398]

The problem of efficiency droop[1,2] in InGaN/GaN quantum well light emitting diodes (LEDs) continues to limit their use in applications that demand a high level of light output at maximum efficiency. Thus there is a great deal of effort worldwide to understand the fundamental causes of this phenomenon, which will then hopefully lead to devices with improved performance. To date the mechanisms that have been suggested to be responsible for efficiency droop are poor efficiency of hole injection,[2–5] Auger recombination,[6,7] carrier escape,[8] carrier loss mechanisms associated with the effects of reduced carrier localisation or saturation of localised states,[9–13] and modification of the free carrier radiative decay constant at high carrier densities.[13] Although the consequence of efficiency droop is particularly important for LED performance at room temperature it has been demonstrated[14,15] that valuable information on the underlying mechanism(s) can be obtained from low temperature measurements. With this in mind we have extended our previous[12] work (which addressed changes in the localization induced S-shape temperature dependence of the peak photoluminescence energy with increasing excitation power density) by investigating the variation of the photoluminescence time decay as a function of excitation power density at a sample temperature of 10 K.

There already exist several reports of such measurements that are of particular relevance to the work discussed here. Shen *et al.*[16] analysed the form of the room temperature decay of the photoluminescence from a relatively thick InGaN layer in terms of the so-called ABC model. In this work analysis of PL decay transients taken at room temperature led to the determination of the C coefficient, attributed to Auger recombination, to be in the range $(1.4\text{–}2.0) \times 10^{-30}$ cm$^6$ s$^{-1}$. Since then values of the Auger coefficient for InGaN quantum wells covering the range $5 \times 10^{-28}$ cm$^6$ s$^{-1}$ to $1 \times 10^{-34}$ cm$^6$ s$^{-1}$ have been determined using a variety of techniques.[17–19] Brosseau *et al.*[20] and Sun *et al.*[21] also reported fast decay transients but attributed their observations to very different mechanisms. Although the main thrust of the work by Brosseau *et al.* in the study of InGaN/GaN quantum wells was a comprehensive analysis of the long lived decay component associated with the decay of localised carriers,[15] they reported for an injection level of $6 \times 10^{12}$ photons cm$^{-2}$ per pulse, at low temperature, recombination on the high energy side of the main PL band with a time constant $<4$ ns which they attributed to free exciton emission. Sun *et al.* also reported a feature in the low temperature PL spectrum with a decay time in the range 1.41–2.22 ns, which they ascribed as due to radiative recombination of carriers in extended states.

In this paper we report on the recombination dynamics and PL spectroscopy of $In_xGa_{1-x}N$/GaN quantum well samples as a function of excitation power density at low temperature (10 K) and, in particular, consider the short-lived decay component observed at high injection densities in the context of a possible mechanism for efficiency droop.

Single quantum well structures were used for the majority of this work as the low temperature PL line width tends to be less than that of multiple quantum well structures since the effects of any well to well variations in well thickness or In fraction are reduced. The structural and optical properties of these single quantum well samples have been reported previously.[22] The samples consisted of a GaN buffer layer 2.5 $\mu$m thick grown on a low-temperature GaN nucleation layer followed by a single 2.5 nm $In_xGa_{1-x}N$ quantum well confined by 7.5 nm-thick GaN barriers. Indium fractions over the range of $x = 0.05$–0.25 were achieved by varying the growth temperature between 800 and 710 °C.

For the optical experiments the samples were mounted on the cold finger of a closed cycle He compressor cryostat. The PL spectra were measured using as an excitation source the chopped output of a mode locked Ti sapphire/pulse picker/frequency doubler system. The characteristics of the excitation radiation were the pulse width was $\sim$100 fs, the maximum time averaged power density was 26 W/cm$^2$, and the reputation rate was 38 kHz. The photon energy of the excitation source varied from sample to sample but was always less than the energy of the GaN bandgap. These excitation

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conditions lead to an estimated maximum carrier density of $\sim 10^{12}$ $cm^2$/per pulse based on an absorption coefficient for InGaN of $10^4\,cm^{-1}$. It has to be stressed that the value of carrier density quoted is very much an estimate as the absorption coefficient will be strongly influenced by the magnitude in the polarisation field and the density of states functions. To avoid the effects of interference[22] on the PL spectra produced by the etalon defined by the GaN/air interface and the GaN/sapphire interface the sample was inclined at Brewster's angle to the collection optics and the light passed through a linear polariser. In this configuration, only that polarisation of light that is not reflected at the GaN/air interface is collected. The photoluminescence emission was focussed onto the slits of a 0.85 m spectrometer and the light detected by a GaAs photomultiplier tube. The time-averaged signal from the photomultiplier was then processed using standard lock-in techniques. The decay characteristics of the emission were measured using time correlated single photon counting. It should be noted that the small peak in the decay curves in Fig. 3 at early times for the large excitation power densities are instrumental in nature.

Of particular importance in this paper is the comparison between the time-integrated PL spectra and the recombination dynamics. We have previously[12] identified two regions of interest in the time integrated PL spectra: (i) an excitation power density regime in which the integrated intensity per unit power (which is directly related to the internal quantum efficiency) remains constant with increasing excitation power density, we term this regime the "plateau region" and (ii) a regime where the integrated intensity per unit power falls as the excitation power density increases (i.e., in which the internal quantum efficiency droops). We have studied these regimes previously at both low (10 K) and room temperature in the work by Hammersley *et al.*[12] An example of this behaviour is shown in Fig. 1 for a single InGaN/GaN quantum well structure at 10 K with a nominal well width of 2.7 nm and an In fraction of 0.15, where up to peak excitation power densities $\sim$100 MW/$cm^2$/pulse the integrated PL intensity per unit excitation power remains constant and then above this critical excitation power density the efficiency falls. For the range of excitation power densities in the plateau region the form of the time integrated PL spectrum remains relatively constant, a typical example is shown in Fig. 2 (dotted line). Also the form of the PL decay curves at different excitation densities in the plateau region remains constant for any particular detection energy. In general the decay curves are non-exponential and can be described by the model put forward by Morel *et al.*[23] where the dynamics can be described in terms of the recombination of independently localised electrons and holes.

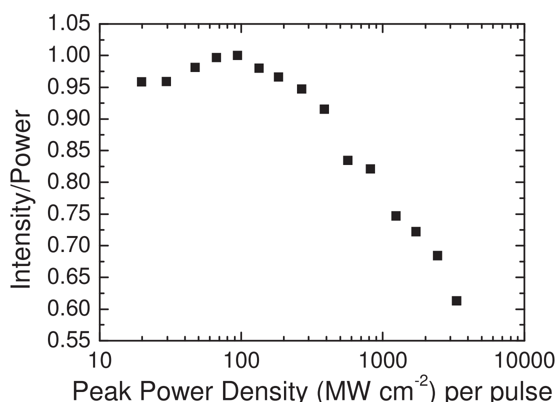


FIG. 1. Plot of normalised integrated PL intensity per unit power at 10 K versus peak excitation power density.

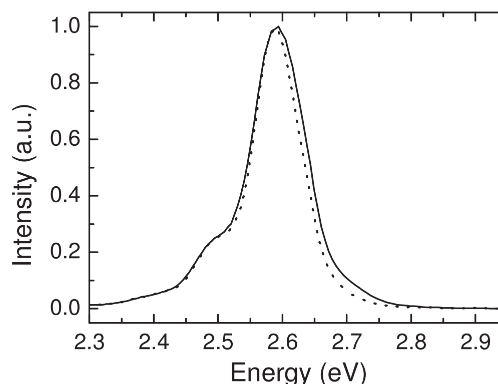


FIG. 2. Normalised time integrated PL spectra for 3330 MW/$cm^2$ per pulse (solid line) and 95 MW/$cm^2$ per pulse (dotted line).

Once the excitation power density is increased into the regime where efficiency droop is observed the time integrated PL spectra and the form of the PL decay curves are modified significantly. In the case of the PL spectrum a distinct broadening on the high energy side of the spectrum is observed as shown in Fig. 2 (solid line). Similar behaviour has been reported most notably in the work reported by Bochkareva *et al.*[13] Along with the emergence of this high energy broadening a significant modification of the decay curves is also observed when we monitor the PL decay on the high energy side of the spectrum as shown in Fig. 3. Once excitation power densities in excess of 700 MW/$cm^2$/pulse are used, a modification to the decay curves at early

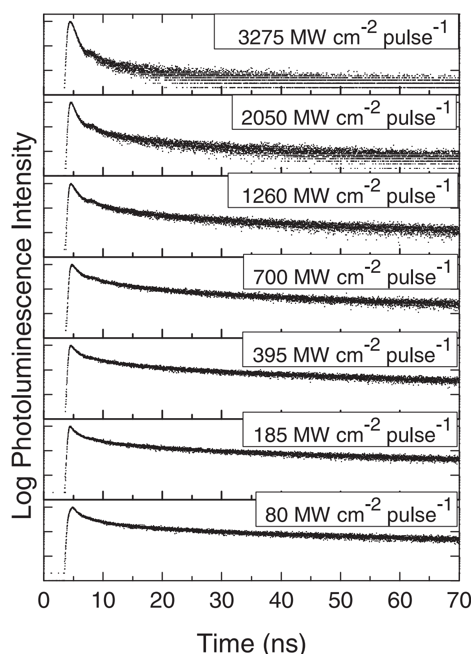


FIG. 3. Photoluminescence time decay curves measured for detection energy of 2.695 eV with the indicated peak power excitation densities per pulse.

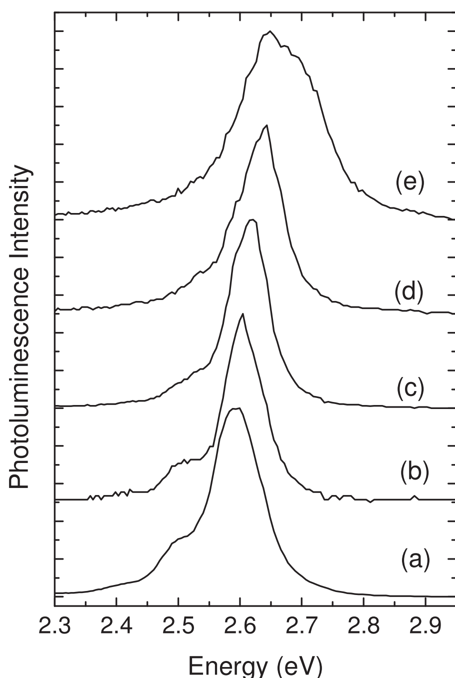


FIG. 4. (a) Time integrated photoluminescence spectrum for an excitation power density of 3640 MW cm$^{-2}$ pulse$^{-1}$ compared with time resolved spectra taken with time windows (b) 17–89 ns, (c) 5–17 ns, (d) 2–5 ns, and (e) 0–2 ns, where zero time is at the peak of the excitation pulse.

times can be seen with the emergence of a fast component. This fast component can be described by a single exponential time constant of 1.9 ns. This suggests that the emission does not involve separately localised electrons and holes and is of a fundamentally different origin. This is born out by the time resolved spectra shown in Figure 4, which compares the time-integrated spectrum (Figure 4(a)) to several time resolved spectra (Figures 4(b)–4(e)). For the detection time windows used for Figures 4(b)–4(d) the general form of the spectra is very similar, the main feature being due to zero phonon emission and at lower energy a shoulder attributable to LO phonon assisted recombination. As the time windows in Figs. 4(b)–4(d) are moved to shorter and shorter time delays the peak energy of the zero phonon emission moves to progressively higher energy. The mechanism responsible for this shift could be either screening of the in-built electric field at earlier times when the instantaneous carrier density is higher or could reflect the change in time scale of the recombination across the emission line that is widely attributed to the effects of changes in the local In fraction. There is some evidence that the electric field is changing due to the simultaneous reduction in the relative strength of the LO phonon assisted recombination.[22] For the earliest time window (Figure 4(e)), a high energy shoulder can be clearly seen at $\sim$2.72 eV with no evidence across the whole spectrum for phonon assisted recombination.

The emergence of this rapidly decaying emission occurs for peak excitation power densities for which we observe bleaching of the S-shape dependence of the peak position as a function of temperature. This disappearance of the S-shape has been explained[12] in terms of saturation of the localised states which prevents the thermal redistribution of carriers amongst the distribution of states. On this basis we attribute the relatively weak, rapidly decaying emission as involving the emission of weakly localised or delocalised carriers. This assignment is similar to that reported previously by Brosseau *et al.*,[20] although it should be noted that it is not possible to say unequivocally whether the measured time constant is dominated by radiative or non radiative recombination.

This leads to an overall picture that at high excitation densities we simultaneously observe a bleaching of the S-shape peak energy shift and the emergence of a rapidly decaying high energy component in the emission spectrum which we ascribe to the recombination of weakly localised or delocalised carriers. This is compatible with the model whereby efficiency droop is explained in terms of carrier loss mechanisms associated with the effects of reduced carrier localisation or saturation of localised states.[9–13]

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