

Photoluminescence measurements on cubic InGaN layers deposited on a SiC substrate

D G Pacheco-Salazar^{1,5}, J R Leite^{1,4}, F Cerdeira², E A Meneses², S F Li³, D J As³ and K Lischka³

¹ Instituto de Física, Universidade de São Paulo, Caixa Postal 66318, São Paulo, SP, Brazil

² Instituto de Física ‘Gleb Wataghin’, Universidade Estadual de Campinas, Caixa Postal 6165, 13083-970 Campinas, SP, Brazil

³ Department of Physics, University of Paderborn, Warburger Strasse 100, D-33098 Paderborn, Germany

E-mail: elieres@ifi.unicamp.br

Received 4 November 2005, in final form 11 April 2006

Published 16 May 2006

Online at stacks.iop.org/SST/21/846

Abstract

In this work we report optical experiments on pseudomorphic cubic $\text{In}_x\text{Ga}_{1-x}\text{N}$ epilayers grown on cubic GaN/3C-SiC templates. We make a detailed study of photoluminescence (PL) and photoluminescence excitation spectroscopy on these samples, with spectra taken at various temperatures (between 2 K and 300 K) and using variable wavelength sources to excite the PL spectra. The combined use of these techniques suggests the existence of indium-rich clusters, constituting a negligibly small fraction of the volume of the total layer. Our results reinforce the notion that the large Stokes-like shift (a difference of approximately 300 meV between emission and absorption) observed in these samples is due to the fact that light absorption occurs in the bulk alloy of average composition while recombination occurs within the indium-rich clusters.

1. Introduction

The growth and device applications of III–V nitride semiconductors experienced fast development after the first successful growth of p-type GaN and the realization of the first blue light emitting diode (LED) in the early 1990s [1]. These LEDs are based on wurtzite (hexagonal) group III nitrides and contained InGaN quantum wells (QW) in the active area. Although these devices are commercially available, the mechanism of light generation is still a subject of ongoing discussion. The fact that InGaN emission is observed at energies far below the energy gap of the alloy which is measured by absorption or ellipsometry and that the high efficiency carrier recombination is insensitive to defects led to the assumption that the photoluminescence (PL) results from excitons localized in In-rich regions [2, 3]. However, in hexagonal group III nitrides piezo-electric and spontaneous

polarization fields give rise to a quantum confined Stark effect (QCSE) if the samples are grown in the (0001) direction (*c*-axis) [4]. This QCSE introduces an additional strong PL red-shift and decorates the phenomena due to localization effects [5]. Alternatively, In segregation may result in the formation of very small $\text{In}_x\text{Ga}_{1-x}\text{N}$ In-rich clusters [6]. These clusters have a smaller energy gap than the bulk alloy and would provide efficient recombination centres. Hence, strong absorption would occur at the energy gap of the bulk alloy (because this occupies the overwhelmingly larger fraction of the volume of the sample), whereas recombination would occur preferentially in the In-rich regions having a smaller energy gap.

In cubic InGaN (*c*-InGaN) epilayers, grown along the (001) direction, strain-induced piezo-electric fields do not exist and there is no associated spontaneous polarization [7]. Therefore several groups have investigated the emission mechanisms in *c*-InGaN on GaAs substrates [8] and on the 7 μm thick 3C-SiC/Si (001) substrates [9] giving evidence that optical processes in InGaN are affected by the localization

⁴ Deceased.

⁵ Permanent address: Dept. de Física, Universidad Nacional de San Agustín Arequipa, Perú.

effects. Micro-Raman and x-ray diffraction of c-InGaN layers grown on GaAs substrates gave first evidences of In-rich inclusions [10] and resonant Raman spectroscopy supports the hypothesis that the PL from these layers is associated with In-rich quantum dot-like structures of nanometre size [11, 12]. It has been argued that, due to the large lattice mismatch between GaN and InGaN, a solid phase miscibility gap in the $\text{In}_x\text{Ga}_{1-x}\text{N}$ alloys exists [6] and spinodal decomposition may take place at the growth temperature leading to the formation of an In-rich phase within the $\text{In}_x\text{Ga}_{1-x}\text{N}$ alloy. However the formation and the size of these In-rich clusters may be influenced by the strain status of the growing $\text{In}_x\text{Ga}_{1-x}\text{N}$ epilayer and a recent paper proposed that strain would even suppress the formation of such phase-separated QDs [13] and acts as a driving force for chemical ordering [14].

The preceding discussion shows that the origin of the large difference between absorption and emission photon energies is not yet settled and more experimental data might throw some light on the subject. In this work we report optical experiments on pseudomorphic c-InGaN epilayers grown on cubic GaN/3C-SiC templates. We make a detailed study of photoluminescence (PL) and photoluminescence excitation spectroscopy (PLE) on these samples, with spectra taken at various temperatures (between 2 K and 300 K) and using variable wavelength sources to excite the PL emission. The technique of PLE combines the properties of both PL and optical absorption. Although the shape of the PLE spectrum mimics the shape of absorption spectrum, the intensities of the different features do not represent actual absorption strengths. Rather, they reflect the effect that such an absorption has on the recombination efficiency of the PL feature on which the analysing spectrometer is centred. Thus, absorption in regions where recombination is very efficient could, in principle, produce features in the PLE spectrum that have considerable intensity, even in the cases where the volume fraction occupied by these regions is so small as to render them undetectable by other techniques [15].

The samples varied in indium molar fraction from 2.8% to 17.1%. The PL spectra of these samples show a main, broad, feature and several sharper structures. By exciting the PL with radiation of progressively lower photon energies (some of which are below the absorption edge of the buffer GaN layer), we identify this main feature of the PL spectrum as being the only one that originates in the alloy layer. Centring the detection on or around this PL feature we take PLE spectra, which reveal the absorption edges of the InGaN layers. These lay consistently some 300 meV above the PL maximum. Reducing the photon energy of the exciting radiation even further, so that now it lies below the absorption edge of the InGaN layer, this main feature in the main PL spectrum still survives. This indicates that this recombination occurs because of absorption occurring within the alloy layer, but not by the ‘bulk’ part of the layer having average composition. This, and the temperature dependence of this PL feature, support the idea that In segregates into clusters of small dimensions. These clusters constitute a negligible fraction of the volume of the InGaN layer, and remain undetected by x-ray diffraction or optical absorption. It is by the combined use of PL and PLE techniques that the presence of this phase is inferred.

Table 1. Description of the cubic $\text{In}_x\text{Ga}_{1-x}\text{N}$ epilayers.

Samples	In molar fraction (x)	Layer thickness (nm)	
		Alloy	GaN buffer
A	0.028	55	540
B	0.065	51	538
C	0.098	40	390
D	0.130	40	439
E	0.171	40	525

2. Experimental details

Cubic $\text{In}_x\text{Ga}_{1-x}\text{N}$ epilayers were grown by plasma-assisted molecular beam epitaxy (MBE) on about 200 μm thick 3C-SiC (001) substrates. A commercial Riber 32 MBE system equipped with an Oxford Applied Research HD 25 radio frequency N-plasma source was used. Conventional Knudsen cells were used for the evaporation of In and Ga. Prior to the growth the 3C-SiC were chemically cleaned and annealed in vacuum at 500 °C for 10 h. Then, a thick cubic GaN buffer layer (about 500 nm) was grown on the SiC substrate at 720 °C [16]. After a growth interruption for reducing the substrate temperature, about 50 nm thick $\text{In}_x\text{Ga}_{1-x}\text{N}$ epilayers were grown at 620 °C. By adjusting the In to Ga flux ratio the In content was varied between $x = 0.028$ and $x = 0.171$. The characteristics of the samples used in our experiments are summarized in table 1. The In content and the strain status were measured by high-resolution x-ray diffraction. Reciprocal space maps of the asymmetric $(-1-13)$ reflection revealed that the thick GaN buffer layers were totally relaxed to the 3C-SiC substrate, whereas all $\text{In}_x\text{Ga}_{1-x}\text{N}$ epilayers were fully strained to the underlying GaN buffer. Full details of the characterization of these, and other, InGaN layers are to be found elsewhere [17].

PL measurements were performed both with laser and arc-lamp excitation. The samples were placed in a JANIS (model SVT-200-5) cold finger cryostat, where the temperature could be varied continuously from approximately 3 K to 300 K. The measurements with arc lamp excitation, both PL and PLE, were performed with an air-cooled 1000 W Xe lamp filtered with a 1.0 m SPEX monochromator with a 1200 groove mm^{-1} grating. The monochromatic light was focused onto the sample, contained in an air products cold finger cryostat at $T = 7$ K. Emitted light was focused into a double monochromator with a Ga—In—As photo multiplier tube. Quartz lenses were used both for excitation and collection of emitted radiation.

3. Results

In figure 1 we show the PL spectra of the InGaN samples listed in table 1. In this figure, PL intensities are displayed in a logarithmic intensity scale. These spectra were taken at 2.5 K using the $\lambda = 325$ nm line of a He—Cd laser as a source of excitation. The spectra were fitted by Gaussian-like peaks and the arrows indicate the maxima of the PL emission for samples with indium molar fraction $x \geq 0.098$. For these samples a strong emission peak around 2.5 eV occurs, which has been previously attributed to recombination at In-rich sites of the sample [8]. As can be seen in figure 1 there is a large shift

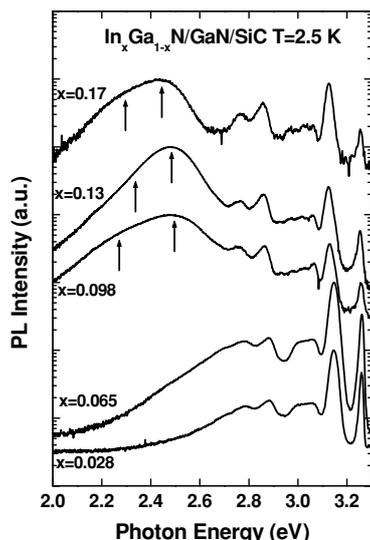


Figure 1. PL spectra taken at 2.5 K, using the 325 nm line of a He–Cd laser in logarithmic intensity scales. In both cases curves have been shifted vertically for the sake of display clarity.

in the PL energy between the layers with $x = 0.028$ – 0.065 and those with $x > 0.098$. This shift may indicate a change of the character of the emission mechanism due to clusters in the disordered phase as discussed in a previous paper [18]. In order to find the roots of the underlying physics we concentrate furthermore on the samples containing higher In content where the emission lines are energetically well separated from the superimposed GaN emission. A secondary maximum at lower photon energies can also be observed in some of the samples (arrows). This signifies that the emission peak is composed of a superposition of peaks centred at different photon energies. In addition, the low-temperature PL spectra of these samples were taken using the filtered radiation from a Xe arc as excitation. The spectra obtained in this way (not shown) are entirely similar to those in figure 1, albeit with a lesser signal-to-noise ratio. All these spectra show a very broad emission at photon energies between 2.5 and 2.7 eV (which may appear as the superposition of more than one peak for some samples) and a number of sharper structures at higher photon energies. Of the latter, some are clearly associated with the emission of the GaN buffer layer (see for instance [8, 15]), while others (in particular sharp lines around 2.9 eV) could be, in principle, also attributed to the InGaN alloy layer.

In order to try to assign correctly the observed PL lines, we proceeded to take low-temperature PL spectra with lamp using progressively higher wavelengths (lower photon energies) as exciting radiation. As the photon energy of the exciting radiation decreases below the absorption edge of the GaN buffer, some lines drop away from the PL spectrum, as illustrated in figure 2(a) for sample D ($x = 0.13$). This figure shows that when the excitation photon energy falls below the GaN energy gap ($h\nu_{\text{exc}} \leq 3.2$ eV), the PL features at 2.75 and 2.85 eV disappear from the spectrum. This identifies these lines as originating in the GaN buffer layer rather than in the InGaN alloy layer. Also some new sharp features appear below 2.3 eV, which lies just below the indirect gap of the 3C-SiC [19]. Since at this low excitation frequencies the light

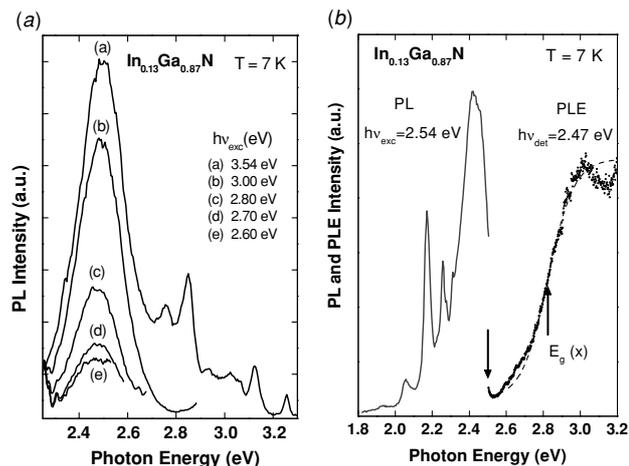


Figure 2. (a) Low-temperature (7 K) PL spectra of sample D obtained by using exciting radiation of progressively lower photon energy and (b) PL spectrum of sample D excited by the 488 nm (2.54 eV) line of an argon laser (solid line) compared to the PLE spectrum of the same sample, centring detection at the maximum of the PL peak (circles), and the fitted absorption profile (dashed line) of the alloy layer obtained as explained in the text.

penetrates all the way into the 3C-SiC substrate, we attribute these new lines to emissions from SiC (bound impurities associated with the indirect gap and their phonon replicas) [20]. Thus, the only feature in the PL spectra of our samples which can be unequivocally attributed to the InGaN layer is the rather broad emission appearing around 2.5 eV for most of our samples. This line survives even for excitations very close to this maximum. This is illustrated in figure 2(b), which shows this PL peak taken with the $\lambda = 488$ nm ($h\omega = 2.54$ eV) line of an argon-ion laser and the PLE absorption profile of the InGaN layer obtained by PLE.

The PLE spectrum (dashed line in figure 2(b)) mimics the absorption spectrum and resembles a broadened step-like function. This lineshape is characteristic of a M_0 singularity with an exciton which is broad enough to merge with the continuum [21]. Fitting this spectrum we obtain a value for the photon energy of the absorption edge, as well as for its broadening parameter Γ . The arrow indicates the location of this critical point in the absorption, which we attribute to the adsorption edge of cubic InGaN. For sample D we estimate a critical point at 2.82 eV and a Γ of 150 meV. In comparison to cubic GaN, where the critical point is located at 3.26 eV and Γ is about 50 meV [15], we observed an increase of Γ with increasing In content (seen also in figure 5). This large width is, in turn, attributed to sample imperfections. The application of a broadened step-like function for the InGaN alloys is even more justified than for the binary GaN since the exciton binding energy is decreasing with increasing In content due to the lower effective carrier masses.

Note that, although the excitation is very close to the emission peak and well below the absorption edge of the alloy layer (indicated by a vertical arrow in the figure), the recombination peak is still clearly observed. This indicates that this recombination occurs because of absorption occurring within the alloy layer, but not by the part of the layer having average composition (henceforward referred to as the bulk

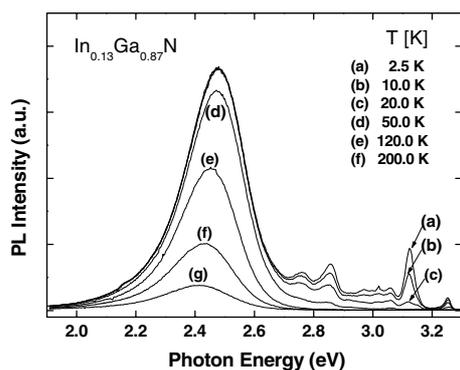


Figure 3. PL spectra of sample D, excited with the 325 nm line of a He–Cd laser, taken at different temperatures.

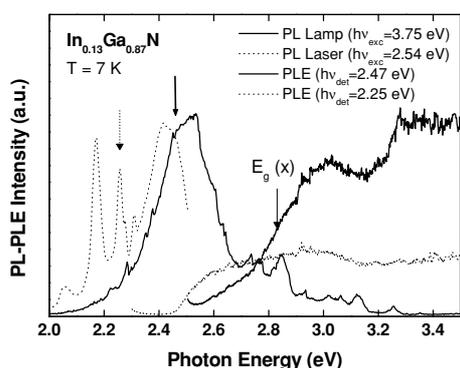


Figure 4. Comparison between PL (obtained by exciting the sample with radiation with two different photon frequencies) and PLE spectra (centred at different emission peaks) for sample D. Dashed lines correspond to PL excited with a photon frequency below the alloy absorption edge and a PLE profile for detection centred at one of the emission peaks of 3C-SiC (dashed arrow), respectively. Solid lines represent PL excited with a photon frequency above the GaN absorption edge and a PLE profile for detection centred at the main emission peak of the alloy layer (solid arrow), respectively.

layer), which has an absorption edge above that of the exciting radiation. Rather, the light appears to be absorbed by the same part of the layer responsible for the emission i.e., the In-rich phase with a lower gap. If this were true, no giant Stokes shift needs to be invoked. Finally, although the whole discussion was based on results from sample D, the same could be argued from the spectra of all the other samples (not shown), which display the same type of behaviour.

Finally, in figure 3 PL shows PL spectra of sample D ($x = 0.13$) taken as a function of temperature, from $T = 2.5$ K up to room temperature. The intensities of PL peaks known to originate in defects, such as the 3C-SiC-related peaks at photon energies at or below 2.3 eV or those related to defects in the GaN layers at energies above 2.6 eV, show the typical exponential decay and become unobservable above $T \approx 80$ K. In contrast, the main PL feature at $E \approx 2.5$ eV persists all the way to room temperature. This behaviour favours the interpretation of phase segregation in the form of In-rich clusters with dimensions small enough to produce a substantial confinement of the carriers trapped in them.

The type of absorption profile depicted in a PLE spectrum depends on the photon energy at which the analysing spectrometer is centred. This is illustrated in figure 4,

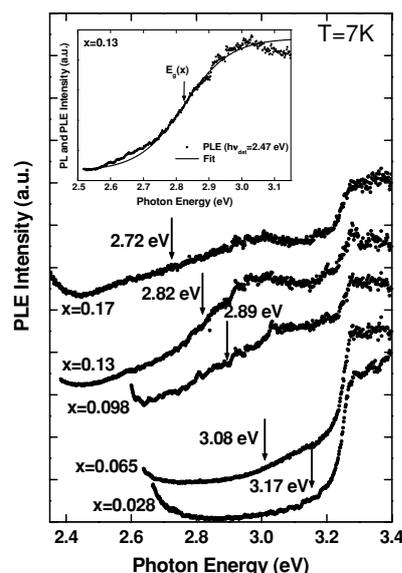


Figure 5. Low temperature (7 K) PLE profiles for detection centred at the main emission peak of the alloy layer. The inset shows an example of the fitting of the PLE spectrum of the same sample with a broadened step function (see text) in order to obtain the absorption profile of the alloy layer.

where the PL and PLE spectra of sample D ($x = 0.13$) are displayed. In this figure, the different PLE spectra are centred at different photon energies. This is indicated by solid or dashed curves. Arrows indicate the centring of the photon energy PL detection for each PLE curve of the same type (dashed or solid). If centring is made around the energy of the PL maximum ($\hbar\omega \approx 2.47$ eV, solid arrow), previously identified as originating during emission from the alloy layer, two clear absorption edges are observed (solid curve): one corresponding to the GaN buffer layer ($E_g = 3.26$ eV) and another at lower photon energy ($E_g = 2.8$ eV, solid arrow), which we attribute to the main absorption edge of the alloy layer. The dashed PLE curve in figure 4 was taken with centring well below the PL maximum ($\hbar\omega \approx 2.25$ eV, dashed arrow) and on the top of PL lines attributed to 3C-SiC emission (see dashed PL curve in figure 4). This curve shows an absorption profile which starts to rise steeply at $E \approx 2.4$ eV (much lower than the absorption edge of the bulk alloy), then saturates, showing a more or less constant absorption, and finally drops again around the photon energy region where the GaN buffer layer starts to absorb light. We attribute this absorption-like feature of the PLE to the 3C-SiC substrate, which has an indirect gap [19, 20] around 2.5 eV. The absorption-like feature saturates and ultimately declines when the absorption of incoming light from the alloy and GaN layers prevents it from reaching the 3C-SiC substrate. Similar results for this lower absorption feature are obtained for all our samples. Since this PLE feature is not related to the properties of the layers we shall discuss it no further.

When the detection frequency is centred around the main InGa_N emission line, the absorption-like features in the spectra have two edges: one corresponding to the gap of the InGa_N layer of average composition and another, around 3.25 eV, corresponding to the gap of the GaN buffer layer. These PLE spectra for all our samples are displayed in figure 5. The arrows

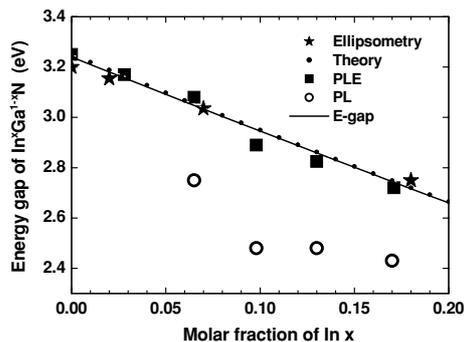


Figure 6. Plot of the dependence of the alloy energy gap versus the In molar fraction (x). Stars represent the ellipsometric data of [22], squares represent our own PLE data and the solid line is a fit of these data to the experimental points using equation (1) in the text. Open circles represent the strongest maxima in the PL spectra.

indicate the alloy absorption edges obtained by assuming that this edge can be fitted by a broadened step function, as is the case in c-GaN thin films [15]. This fitting is again illustrated in the inset of this figure for the case of sample D ($x = 0.13$).

Finally let us turn to the absorption edge of the bulk InGaN layers obtained from figure 5. A plot of $E_g(x)$ versus x is shown in figure 6, where values obtained by ellipsometry [22] in c-InGaN layers deposited on GaAs are also plotted for comparison purposes. A good agreement exists between our PLE data and the ellipsometric data obtained by other authors on cubic InGaN samples grown on GaAs substrates. The slight energy difference between both sets of data (~ 72 meV) is due to the fact that our PLE data are taken at 7 K while the ellipsometry data were taken at room temperature. In this figure we also plot the results of $E_g(x)$ obtained by Teles *et al* [23] (dotted curve), who use an energy gap of InN as $E_g(\text{InN}) = 0.9$ eV [24] and a parameter-free calculation of electronic properties of $\text{In}_x\text{Ga}_{1-x}\text{N}$ alloys based on a generalized quasi-chemical approach and a pseudopotential plane-wave method. Recent experimental data by Wu *et al* [25] and Matsuoka *et al* [26] indicate that the band gap of hexagonal InN is at about 0.7 eV. Since the band gap of cubic InN shall be smaller than that of hexagonal InN we assume a fundamental gap of about 0.6–0.7 eV for the zincblende phase [27]. By using the commonly applied expression

$$E_g(x) = (1-x)E_g(\text{GaN}) + xE_g(\text{InN}) - bx(1-x), \quad (1)$$

we estimated bowing parameters b of 0.4 eV and 0.45 eV for band gaps of 0.6 eV and 0.7 eV, respectively. In figure 6 the solid curve was obtained using a bowing parameter of $b = 0.4$ eV and $E_g(\text{InN}) = 0.6$ eV. How does this gap $E_g(x)$ relate to the emission line for each of these samples? The positions of these emission peaks are also plotted in figure 6 as open circles. The experimental data indicate that a shift of more than 300 meV seems to exist between absorption and emission for all our samples. A closer look reveals that the recombination energies, corresponding to the higher energy peak of the broad PL feature originating in the alloy layer, are only very weakly dependent on the average In molar fraction, while the absorption edge follows the relationship between E_g and x given by equation (1). A similar behaviour is exhibited by the emission peaks of samples containing double heterostructures

of InGaN with different In molar fractions [18], in which two PL peaks are present and these have a weak dependence on x . This suggests that the composition of the In-rich clusters is rather insensitive to changes of the average composition of the layer.

4. Discussion

The optical data discussed in the previous section have definite information as to the structure and composition of the InGaN layers. First, the PL spectrum reveals a broad structure (figure 1) which is the only feature of the observed spectrum that can unequivocally be attributed to recombination processes taking place within the alloy layer of our samples (figure 2(a)). This feature is not only broad, but in some cases shows secondary maxima (figure 1). Also, this feature lies well below that of the absorption edge of the layer and can be excited by radiation of photon energies lying well below this absorption edge. In fact, a look at figure 2(b) shows that even when this photon energy is located at points where the absorption of the InGaN layer is zero (dashed line in this figure), this feature still appears in the spectrum. This behaviour clearly indicates that light is being absorbed within the InGaN layer, but it is not the bulk layer of average composition that is responsible for this absorption. Rather, the light appears to be absorbed by the same part of the layer responsible for the emission. This could, in principle, be identified as an indium-rich phase (with a lower gap) occupying a negligible fraction of the layer's volume. The smallness of this fraction would be the reason why the existence of this phase goes undetected by ordinary absorption (or absorption-related techniques such as ellipsometry) and x-ray diffraction. In this case, the energies for both emission and absorption of light would be quite close and no giant Stokes shift needs to be invoked. In order to distinguish between this explanation and a competing one of excitons localized in In-rich regions [2, 3], we studied the temperature behaviour of the PL intensity (figure 3). Any exciton-like feature would show an exponentially decaying intensity as temperature increases, as indeed features in the PL spectra of our samples known to have this origin show. The latter disappear from the PL spectrum for temperatures above $T \approx 80$ K. In contrast, the main PL feature under discussion persists all the way to room temperature. This behaviour favours the interpretation of phase segregation in the form of In-rich clusters with dimensions small enough to produce a substantial confinement of the carriers trapped in them. Also, the fact that the PL peak is broad and appears to have more than one maximum can be attributed to the existence of In-rich clusters of different sizes (with consequently different confining energies) or compositions. But, if our interpretation is correct, why cannot we observe an absorption profile in our PL spectra that corresponds to the absorption of these In-rich clusters? The reason is that this absorption coincides with the photon energy region where the 3C-SiC substrate also absorbs and is masked by the absorption of this overwhelmingly larger part of the complete sample (see the discussion associated with figures 4 and 5).

The PLE spectra of our samples allow the determination of the energy gap, $E_g(x)$, of the bulk alloy of average composition.

The dependence of $E_g(x)$ on x agrees with that obtained by other authors on cubic InGaN samples grown on GaAs substrates [21], and is well fitted with a quadratic function for $E_g(x)$ that uses values for the bowing parameter and the InN gap in good agreement with current estimates for these parameters. The position of this energy gap is consistently higher than that of the emission peak in the PL spectrum. The latter, however, seems to vary very little from the average alloy composition, in consonance with recent data from other InGaN nanostructures [18].

We can only speculate as to the reason for this behaviour. Teles *et al* [14] have calculated the energetic and thermodynamic properties of cubic $\text{In}_x\text{Ga}_{1-x}\text{N}$ using first principles total energy calculations and Monte Carlo simulations. They found that the formation of ordered phases of fixed compositions is energetically favourable in cubic InGaN under biaxial strain. Some of the observed luminescence may be due to recombination of electron–hole pairs in clusters of these phases, since the higher photon energy emission is close to the band gap of $\text{c-In}_{0.25}\text{Ga}_{0.75}\text{N}$, the $[\text{InN}]_1[\text{GaN}]_3$ ordered phase. Thus, the higher emission may be due to the band gap of the ordered phase whereas the lower energy PL peak might correspond to other ordered phases, may be $[\text{InN}]_2[\text{GaN}]_3$ or $[\text{InN}]_3[\text{GaN}]_3$. In any case, the existence of segregated phases in fully strained samples seems to indicate that the formation process is not necessarily due to a bulk spinodal decomposition, which was assumed in the calculations in [13].

5. Conclusions

We make a detailed study of PL and PLE on pseudomorphic c-InGaN epilayers grown on cubic GaN/3C-SiC templates. PL spectra were taken both as a function of temperature ($2.5 \text{ K} \leq T \leq 300 \text{ K}$) and the wavelength of the exciting radiation. Our data are consistent with the existence of In-rich clusters which constitute a negligible fraction of the volume of the total layer and are, therefore, not detected by other optical techniques or even by x-ray diffraction. Nevertheless, because the recombination efficiency within the clusters is so large, this In-rich phase dominates the PL spectrum. In contrast, absorption is dominated by the majority phase, of average composition and consequently larger energy gap. Thus, the rather large energy difference between the PL maximum and the absorption edge in a given sample is due to the difference between the energy gaps of the two phases. Finally, the recombination energies are only very weakly dependent on the average In molar fraction, while the absorption edge follows the relationship between E_g and x given by equation (1). This suggests that the composition of the In-rich clusters is rather insensitive to changes of the average composition of the layer, which might be related to the presence of order in the segregated phases.

Acknowledgments

The authors are thankful to Fundação de Amparo à Pesquisa do Estado de São Paulo (FAPESP, Brazil) and Conselho

Nacional de Desenvolvimento Científico e Tecnológico (CNPq- Brazil) for funding in the forms of research grants and fellowships. Financial support is gratefully acknowledged to Deutsche Forschungsgemeinschaft (DFG-Germany) and the German–Brazilian academic exchange program of DAAD-PROBRAL.

References

- [1] Nakamura S 1991 *Japan. J. Appl. Phys.* **30** L1075
- [2] Chichibu S F *et al* 2000 *Appl. Phys. Lett.* **76** 1671
- [3] Bellaiche L, Mattila T, Wang L W, Wei S H and Zunger A 1999 *Appl. Phys. Lett.* **74** 1842
- [4] Bernardi F and Fiorentini V 1998 *Phys. Rev. B* **57** R9427
- [5] Wetzel C, Takeuchi T, Amano H and Akasaki I 1999 *J. Appl. Phys.* **85** 3786
- [6] Teles L K, Furtmueller J, Scolfaro L M R, Leite J R and Bechstedt F 2000 *Phys. Rev. B* **62** 2475
- [7] Chichibu S F *et al* 2003 *J. Vac. Sci. Technol. B* **21** 1856
- [8] 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
- [9] Chichibu S F *et al* 2001 *Appl. Phys. Lett.* **79** 3600
- [10] Silveira E, Tabata A, Leite J R, Trentin R, Lemos V, Frey T, As D J, Schikora D and Lischka K 1999 *Appl. Phys. Lett.* **75** 3602
- [11] Lemos V, Silveira E, Leite J R, Tabata A, Trentin R, Scolfaro L M R, Frey T, As D J, Schikora D and Lischka K 2000 *Phys. Rev. Lett.* **84** 3666
- [12] Husberg O, Khartchenko A, Vogelsang H, As D J, Lischka K, Noriega O C, Tabata A, Scolfaro L M R and Leite J R 2002 *Physica E* **13** 1090
- [13] Tabata A, Teles L K, Scolfaro L M R, Leite J R, Kharchenko A, Frey T, As D J, Schikora D and Lischka K 2002 *Appl. Phys. Lett.* **80** 769
- [14] Teles L K, Ferreira L G, Leite J R, Kharchenko A, Husberg O, As D J, Schikora D and Lischka K 2003 *Appl. Phys. Lett.* **82** 4274
- [15] Fernandez J R L, Noriega O C, Soares J A N T, Cerdeira F, Meneses E A, Leite J R, As D J, Schikora D and Lischka K 2003 *Solid State Commun.* **125** 205
- [16] Schikora D, Hankeln M, As D J, Lischka K, Litz T, Waag A, Buhrow T and Henneberger F 1996 *Phys. Rev. B* **54** R8381
- [17] Pacheco-Salazar D G, Cerdeira F, Meneses E A, Leite J R, Li S F, As D J and Lischka K 2005 *J. Cryst. Growth* **284** 379–87
- [18] Li S F, As D J, Lischka K, Pacheco-Salazar D G, Scolfaro L M R, Cerdeira F and Meneses E A 2005 *Mater. Res. Symp. Proc.* **831** E8.15
- [19] Harris G L (ed) 1995 *Properties of Silicon Carbide* (London: INSPEC) p 31
- [20] Yamada T and Itoh K M 2002 *Mater. Sci. Forum* **389–393** 675
- [21] Aspnes D E 1980 *Handbook on Semiconductors* ed T S Moss (Amsterdam: North-Holland) p 109
- [22] Goldhahn R, Scheiner J, Shokhovets S, Frey T, Kohler U, As D J and Lischka K 2000 *Appl. Phys. Lett.* **76** 291
- [23] Teles L K, Furtmueller J, Scolfaro L M R, Leite J R and Bechstedt F 2001 *Phys. Rev. B* **63** 85204
- [24] Davydov V Yu *et al* 2002 *Phys. Status Solidi b* **229** R1
- [24] Davydov V Yu *et al* 2002 *Phys. Status Solidi b* **229** R4
- [25] Wu J, Walukiewicz W, Shan W, Yu K M, Ager J A III, Li S X, Haller E E, Lu H and Schaff W J 2003 *J. Appl. Phys.* **94** 4457
- [26] Matsuoka T, Okamoto H, Nakao M, Harima H and Kurimoto E 2002 *Appl. Phys. Lett.* **81** 1246
- [27] Bechstedt F and Furthmüller J 2002 *J. Cryst. Growth* **246** 315