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Composition dependence of photoluminescence properties of $In_xAl_{1-x}N$/AlGaN quantum wells

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A series of InAlN/AlGaN five quantum well (QW) heterostructures was prepared by metal-organic vapour phase epitaxy to investigate their photoluminescence (PL) properties as a function of indium content in QWs at aluminium content in barriers fixed at 59%. In addition to the expected redshift of the emission spectrum, a strong rise of PL efficiency was observed with increasing indium content from 12.5 to 18%. Use of a higher indium content leads to a further redshift but also to a sudden and sharp degradation of PL efficiency. Reasons for the observed behaviour are discussed in detail, which raise the possibility of a transition to a type II band lineup in the InAlN-AlGaN system.

1. Introduction

Besides the large range of band-gap potentially coverable by InAlN compounds, they have attracted the attention of many researchers over last decade due to the potential of lattice matching to GaN with good contrast in refractive indices and spontaneous polarisation constants. This has led to two main areas of current interest: Bragg reflectors [1] and high electron mobility transistors [2]. Some other applications such as using InAlN as electron blocking layers in light emitting diodes (LED) [3], barriers for GaN/InAlN quantum wells (QW) [4] or in UV photodiodes [5] have been also proposed. Despite the

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idea to use InAlN in ultraviolet (UV) LEDs inspired by success of InGaN for visible LEDs, until recently there have been no reports on the use of InAlN as an active region material for light emitting heterostructures. In our recent work, it was shown that strong room temperature (RT) photoluminescence (PL) can be achieved from InAlN QWs sandwiched between AlGaN barriers using quasi-2T growth of the active region, using deposition of a low temperature AlGaN cap layer to prevent QW In-desorption during the ramp to high temperature (1110°C) at which majority of the AlGaN barriers are grown [6]. Additionally, optimal QW width ranges were found to be 1.5-2 nm for QWs grown at 730°C and 2-3 nm for those grown at 790°C. Here we report on systematic study of PL properties of ~2 nm thick $In_xAl_{1-x}N/Al_{0.59}Ga_{0.41}N$ MQWs as a function of indium content.

2. Methods

InAlN/AlGaN 5QW samples studied here were grown by metalorganic chemical vapour deposition (MOCVD) in an AIXTRON 3x2" vertical flow showerhead-type MOCVD reactor. Trimethylindium, trimethylaluminium and trimethylgallium were used as precursors for group III metals, and ammonia as a source of nitrogen. Growth of the active region in N_2-ambient was preceded by deposition of a 2 μm thick undoped AlGaN-buffer layer on pre-prepared AlN/sapphire-templates. More details on preparation of both AlN-template and QW stack on AlGaN buffer are presented elsewhere [6, 7]. Indium content in QWs was varied by changing QW growth temperature T_{QW} in the 710-790°C range while keeping barrier growth temperature fixed at 1110°C. The QW growth time was fixed to give 2 nm well width based on a thick InAlN layer, and the variation of InAlN growth rate was estimated to be ±5% across the temperature range studied. To protect QW from indium desorption during the temperature ramp, an AlGaN cap layer of optimised thickness was grown before starting the ramp, as described in [6]. X-ray diffraction (XRD) was applied to estimate QW indium content using a PANalytical X'pert PRO XRD system in combination with analysis of calibration layers for AlGaN composition. PL properties were studied by exciting the InAlN-based QW samples with the second harmonic of an Argon ion laser (cw, 244 nm) and detecting the emission with a Horiba *i*HR320 imaging spectrometer equipped with Synapse® thermoelectrically cooled CCD detector. The same spectrometer equipped also with a photomultiplier was used for PL excitation (PLE) measurements while luminescence was excited by emission of a Xe lamp dispersed using a double grating monochromator (Horiba Gemini 180). For temperature resolved PL measurements, samples were placed into a Janis Research closed cycle refrigerator system CCS-400/202.

3. Results and discussion

To determine the temperature dependence of indium content in the QWs of our samples, XRD ω-2Θ experimental data was modelled using dynamic X-ray scattering theory [8] based on a commercial package, taking into account known (from separate calibration runs) compositions and thicknesses of buffer and barriers (see Fig. 1 for an example). The resulting indium content (determined assuming MQW stack fully strained to the buffer) was found to increase linearly as the temperature decreases from 790°C down to 730°C (square data points in Fig. 2). Further decrease of temperature led to some apparent saturation of QW indium content but continuation of the linear trend could be achieved assuming a QW relaxation degree of about 1.5% per well (circular data points in Fig. 2).

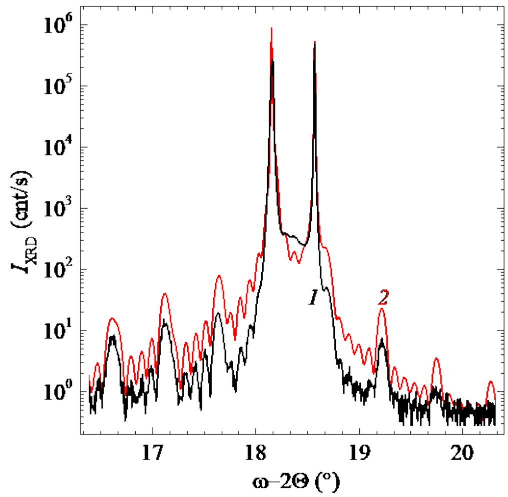

Fig. 1. – 0002 ω-2Θ XRD scan of the InAlN/AlGaN MQW heterostructure grown at T_{QW} = 730°C: *1* – experiment, *2* – model.

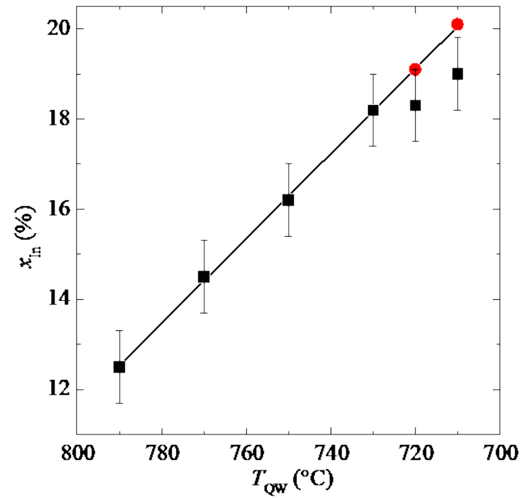

Fig. 2. – QW indium content as a function of T_{QW} determined from XRD data assuming fully strained (squares) and partly relaxed (circles) QWs.

Unfortunately, it was not possible to confirm the exact strain state of samples' MQW stacks by measuring their 105 reciprocal space maps due to absence of MQW-related peaks in the corresponding scans. In our recent work [9] we showed 80-120 nm thick $In_xAl_{1-x}N$ starts to relax at $x \approx 0.11$ when grown on an AlN template. Here the InAlN QWs are only ~2 nm thick and are grown on a partly (10-30%) relaxed AlGaN buffer, therefore, it is not surprising some initial relaxation may occur at higher indium composition.

Room temperature (RT) PL spectra of the InAlN/AlGaN MQW samples are presented in Fig. 3, *a*. The origin of the observed PL bands was determined in our previous work using PL excitation spectroscopy [6]. The weak shortest wavelength band at around 4.75 eV corresponds to the band-edge emission from AlGaN bulk buffer. A somewhat broader and in some cases dominant near UV band is the emission from QWs. Two broad peaks in the visible are defect related emission bands from AlGaN layers.

One can see from Fig. 3, *a* that the QW-related PL band redshifts with decreasing QW growth temperature, which is consistent with the higher indium incorporation and thus lower band-gap of the resulting InAlN. Taking into account In compositions given with circles in Fig. 2 (i.e. determined assuming a partial InAlN relaxation for samples grown at two lowest temperatures), the overall PL redshift is nearly linear with x and is of about 70 meV per one percent of composition (Fig. 3, *b*).

The composition dependence of PL FWHM is presented in Fig. 3, *b* (circles). Unlike InGaN QWs, which typically demonstrate an increasing PL bandwidth with indium composition [10, 11] due to enhanced material inhomogeneity and thus band-gap fluctuation, our InAlN QWs show an opposite behaviour up to about 18% of indium. Only a further increase of indium content seemingly leads to the expected PL band broadening, though the differences in FWHM in this region are fairly small. The reason for the observed behaviour of PL FWHM is most probably related to the strong dependence of PL efficiency on PL peak position (through indium content).

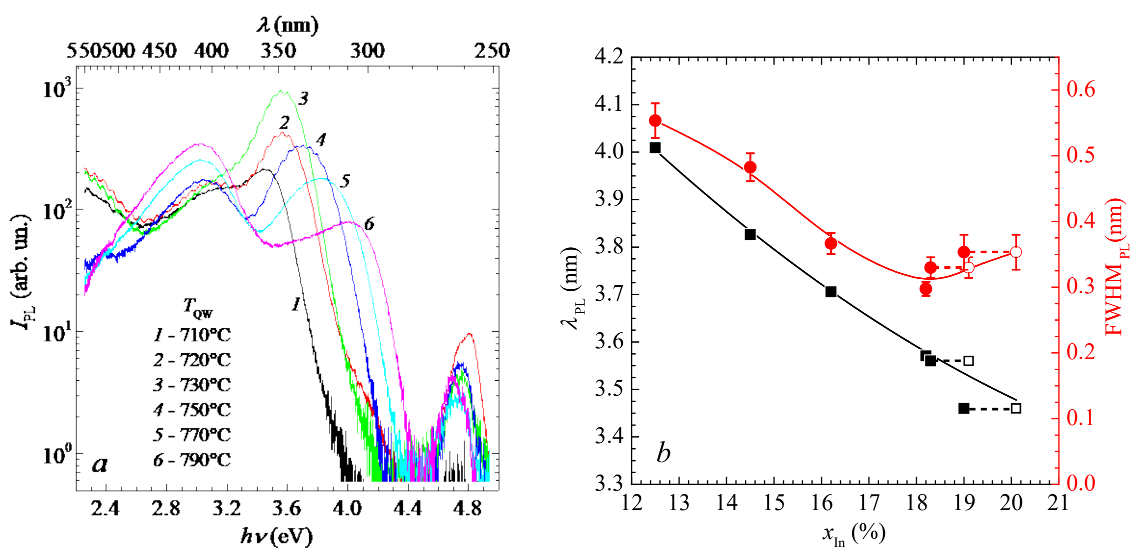

Fig. 3. – PL spectra of InAlN/AlGaN MQW samples grown at different QW temperatures (*a*) and PL peak position (squares) and bandwidth (circles) as a function of indium content (*b*). Open symbols

are plotted for two samples grown at lowest temperatures using x values estimated assuming partial InAlN relaxation (circles in Fig. 2). Curves are guides to the eye.

There are two notable features of the indium content dependence of PL efficiency of the studied samples, which can be seen in Fig. 4: The highest PL efficiency is for $In_xAl_{1-x}N$ QWs with $x \sim 0.18$ with a gradual decrease with reducing In content and a more sudden fall at higher contents. The rapid decrease of PL efficiency for $x > 0.18$ correlates with the point where the rate of change in determined In content (assuming fully strained MQWs) with growth temperature alters, as shown in Fig. 2, which we have attributed to the onset in plastic deformation in the QWs. Such plastic deformation (dislocation formation within the QW structure) leads to reduction in optical efficiency. It is worth noting that we have observed no significant correlation of the MQW-related PL with the crystalline quality of AlGaN/AlN-templates in any study we have conducted in this system. However new dislocations generated within QW material are very critical from this point of view. This is because the threading dislocations intersecting QW plane from the buffer "corrupt" seemingly arbitrary regions of QW and not necessarily In-rich ones, leaving a lot of them dislocation-free where carriers can be localised and recombine radiatively. On the other hand QW based misfit dislocations happen in the most strained and thus most indium rich regions of those QWs. These dislocated In-rich regions are still good traps for non-equilibrium carriers but unfortunately being localised there they have obviously a much lower chance of radiative recombination. We thus conclude that the onset of mechanical strain relaxation during growth of InAlN/AlGaN MQW stack is the main reason of the rapid PL degradation at $x > 18\%$. It is worth noting that in our previous work [6] we observed a decrease of PL efficiency due to the same mechanism at two fixed indium compositions when QW widths were increased instead. The mechanical strain relaxation via misfit dislocation formation occurred at above critical QW widths of about 2 nm and 3 nm for samples grown at 730°C and 790°C respectively.

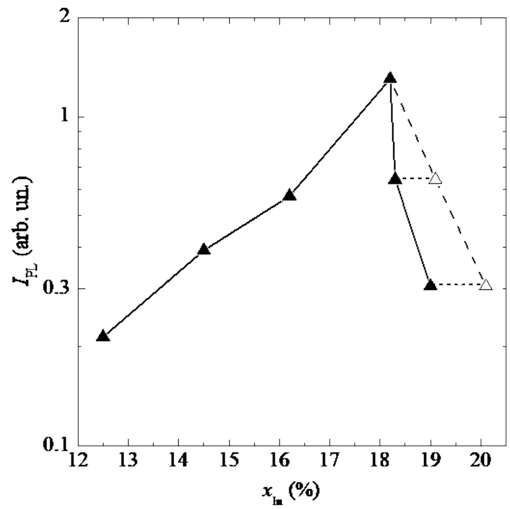

Fig. 4. – Relative PL efficiency of InAlN/AlGaN MQW samples grown at different QW temperatures as a function of indium content. Open symbol data points are plotted in the same way as explained for Fig. 3, *b*.

Let us now consider the gradual decrease of PL efficiency as x falls from 0.18 to 0.12 in our material. Such a behaviour has been observed in InGaN [12, 13] where a reduction in luminescence efficiency with decreasing In content was explained by carriers being localised away from defects in In-rich regions. This behaviour was observed however for InGaN "with an In composition of nearly zero" [12], and for moderate indium contents of about 10% and higher the tendency is usually the opposite [10, 14]. Moreover this initial increase of PL efficiency in InAlN has been observed by our group to happen for indium contents in the range $0< x < 0.01$ [9]. This is consistent with much stronger expected carrier localisation in InAlN in comparison to InGaN due to the higher difference in band-gap and especially due to very high band-gap bowing parameter (> 25 in the $x \rightarrow 0$ limit [9]) which being combined together make the value of dE_g/dx exceptionally high in this content range. A high dE_g/dx will result in strong band-gap variations even for relatively low fluctuations of alloy composition. It is thus clear that relatively low indium content InAlN can generate sufficient localised states to suppress nonradiative recombination, and a further increase in localised state density should have little further benefit. Therefore the deterioration of PL efficiency observed in this study requires a different explanation.

As the QW band-gap gets closer to that of the barriers, obviously carrier confinement within them becomes weaker. For the 12.5% InAlN sample, the QW band-gap (4.38 eV) is still significantly lower than that of their barriers (4.74 eV). However, it is possible in the $In_xAl_{1-x}N$-$Al_yGa_{1-y}N$ system that a type II band lineup can occur, whereby the holes are confined to the $Al_yGa_{1-y}N$ while the electrons are confined as desired in $In_xAl_{1-x}N$. A simple calculation taking into account InAlN and AlGaN band-gap composition dependencies and band offset ratios $\Delta E_{\mathrm{c}}/\Delta E_{\mathrm{v}}$ of 1.33 for the AlN-GaN system [15] and 2.63 for the AlN-InN system [16] indicates a transition from type I $In_xAl_{1-x}N/Al_{0.59}Ga_{0.41}N$ QWs to a type II lineup at $x_{\mathrm{tr}} \approx 0.15$: i.e. for $x > x_{\mathrm{tr}}$ both $E_{\mathrm{cbm}}^{\mathrm{InAlN}} < E_{\mathrm{cbm}}^{\mathrm{AlGaN}}$ and $E_{\mathrm{vbm}}^{\mathrm{InAlN}} > E_{\mathrm{vbm}}^{\mathrm{AlGaN}}$, and thus both electrons and holes are confined in InAlN QW, but for $x < x_{\mathrm{tr}}$, $E_{\mathrm{cbm}}^{\mathrm{InAlN}} < E_{\mathrm{cbm}}^{\mathrm{AlGaN}}$ and $E_{\mathrm{vbm}}^{\mathrm{InAlN}} < E_{\mathrm{vbm}}^{\mathrm{AlGaN}}$ giving AlGaN barrier hole confinement. (The indexes "cbm" and "vbm" stand for conduction band minimum and valence band maximum, respectively). The type II band alignment should result in decreased electron and hole wave function overlap and hence a reduced probability of radiative recombination of the spatially separated carriers. This in its turn leads to reduced efficiency of luminescence if there is a nonradiative recombination channel that is (relatively) unaffected by the carrier separation. It should be noted that both electrons and holes confined in InAlN are likely to spend much time in localised states and not diffuse to nonradiative centres. However in the case of holes confined in AlGaN barriers, they may be freer to move from state to state due to the much weaker potential fluctuations in AlGaN and have therefore a higher chance to be captured by nonradiative and radiative centres. These reasons may explain both the observed reduction of the MQW-related PL band efficiency with decreasing indium content and its anti-correlation with the defect-related PL band efficiency at about 405 nm (Fig. 3, *a*).

To further investigate the possibility of the type I–type II transition, we have undertaken temperature dependent PL on four samples with estimated $In_xAl_{1-x}N$ QW compositions of $x = 0.182$, 0.162, 0.145, 0.125, respectively. These spectra are shown in Fig. 5, *a*-*d*. For $x = 0.182$ (Fig 5, *a*) a fairly simple increase in QW intensity is observed as the temperature is reduced from 450K to 11K. The QW related peak has a shape well described by a single band and for which the small wavelength shifts are consistent with a standard carrier localisation model (not shown). However for the $x = 0.162$ sample (Fig 5, *b*) a distinct apparent blue shift in the QW luminescence is observed as the temperature falls to 11 K. This "blue shift" is best accounted for by the addition of a second band at slightly higher energy which increases more rapidly with decreasing temperature. The validity of such an approach can be confirmed by examination of the temperature dependence of the spectra for $x = 0.145$ (Fig 5, *c*) where the observed behaviour is stronger so that the high energy shoulder becomes dominant at 11 K. The data for the final sample in the series ($x = 0.125$, Fig 5, *d*) is somewhat compromised by the AlGaN buffer donor-acceptor pair band and shows other features beyond the scope of this study, but the main QW peak again appears to show an intensity increase on the high energy side of the band.

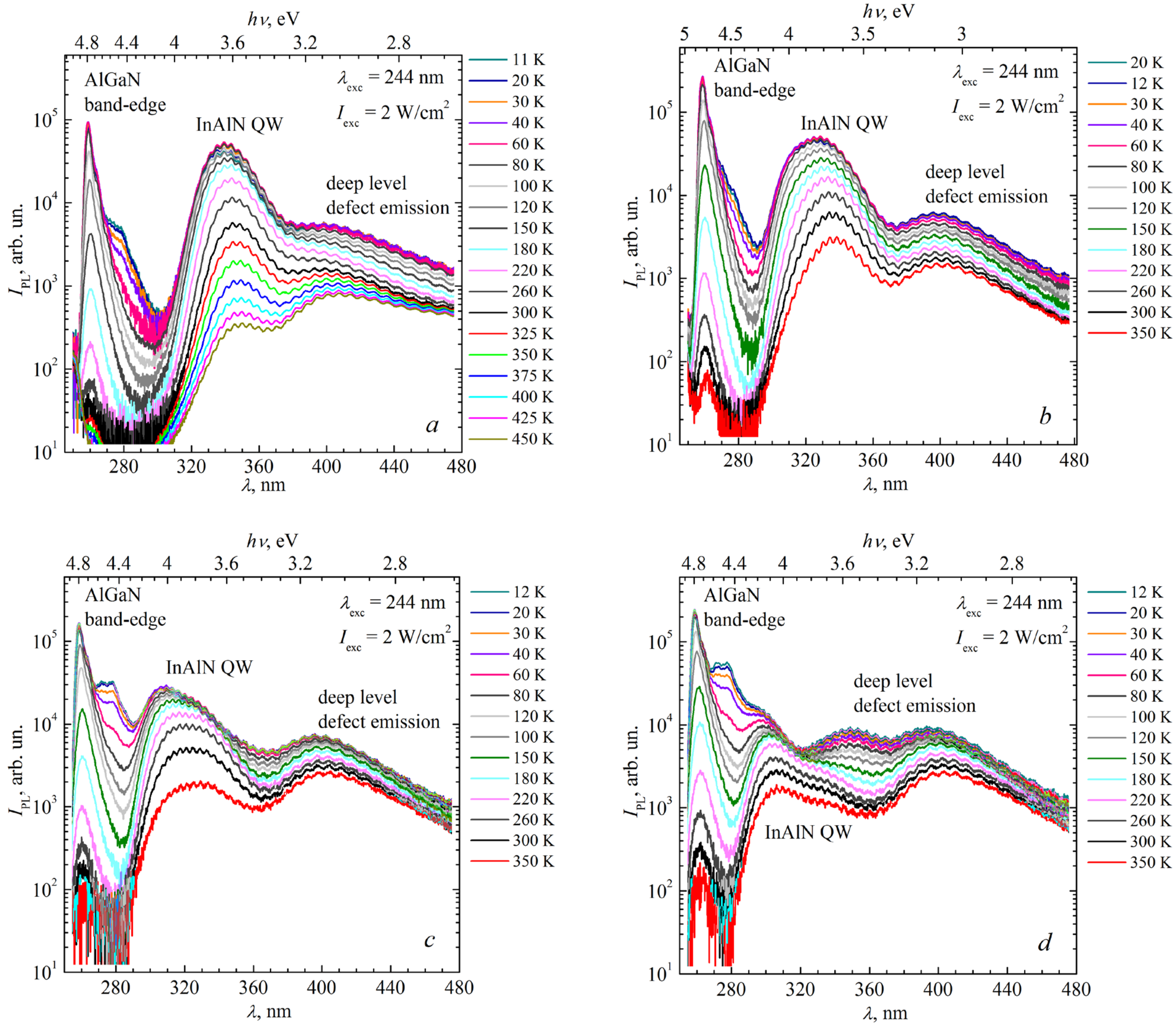

Fig. 5. – Temperature resolved PL spectra of the InAlN/AlGaN QW samples with *a* – *x* = 0.182, *b* – 0.162, *c* – 0.145, *d* – 0.125.

One explanation for the observed behaviour is that there is a competition between two radiative recombination pathways: one with holes in deeply localised centres within the InAlN, in regions of locally relatively high In content; and the other with holes localised in the AlGaN. The holes in AlGaN at room temperature have the ability to more easily "hop" to non-radiative sites (as is typical for AlGaN where PL efficiency drops quickly for the average threading dislocation densities are as high as mid-10^9 cm^{-2} in our templates). Thus the high energy shoulder observed at low temperature for lower values of x may be attributed to recombination from the type II band lineup, while the band which dominates at room temperature is related to that from type I regions of QW (with locally higher indium content).

Having considered the observed degradation of RT PL efficiency with decreasing indium content and temperature dependent behaviour of PL, it is now easier to understand the rather unexpected dependence of PL FWHM (Fig. 3, *b*). For this let us consider the universal relation between absorption $k(\varepsilon)$ and luminescence $I_L(\varepsilon)$ spectra in semiconductors [17]:

$$\frac{I_L(\varepsilon)}{\varepsilon^3 k(\varepsilon)} = \frac{4}{c^2} \cdot \frac{1 - e^{-\Delta F/kT}}{1 - e^{-\varepsilon/kT}} \cdot \frac{1}{e^{(\varepsilon - \Delta F)/kT} - 1}, \tag{1}$$

where $\varepsilon = h\nu$ is the photon energy, ΔF is the distance between quasi Fermi levels for electrons and holes, c is the speed of light in vacuum, and kT is the thermal energy. Normally for wide band-gap semiconductors under optical excitation both $\Delta F \gg kT$ and $h\nu \gg kT$. Moreover the low energy tail of band-edge emission corresponds to the photon energies that are much less than the difference between quasi Fermi levels in comparison to the thermal energy. In this condition $e^{-\Delta F/kT} \approx 0$, $e^{-\varepsilon/kT} \approx 0$ and $e^{(\varepsilon - \Delta F)/kT} \approx 0$ too, which allows simplification of Eq. (1) to:

$$\frac{I_L(\varepsilon)}{\varepsilon^3 k(\varepsilon)} = const \tag{2}$$

i.e. the low energy tail of band-edge emission in the excited material should be proportional to that of the absorption spectrum multiplied by the photon energy to the power of three. This is easy to understand assuming PL as a product of the joint density of states function, matrix element of transition and carrier distribution functions. For the low energy tail of luminescence spectrum, the corresponding states in the conduction band are almost fully occupied and those in the valence band are almost fully free thus resulting in carrier distribution functions that are nearly equal to one. For non-homogeneous materials like III-nitride alloys, especially indium containing ones, the band-gap cannot be given with a single figure for the whole crystal but it is rather a function of coordinates. This leads to broadening of both absorption and emission spectrum, but Eq. (2) normally remains valid as one can see from Fig. 6. The slope of low energy wing of MQW-related band in the PL spectrum of sample grown at 730°C represented in semi-logarithmic scale is nearly equal to that of its PLE absorption edge (both are shown with arrows in Fig. 6). The broadening parameter ΔE calculated for PLE spectra of all samples using a standard sigmoidal expression [18] decreases with increasing QW growth temperature (Fig. 7) showing the expected lowering in the degree of inhomogeneity with decreasing average indium content. This means that the slope of $\ln\left(\varepsilon^3 k(\varepsilon)\right)$ decreases as well. However, one can see from Fig. 3, *a* that the slope of the low energy wing of PL spectra changes in the opposite way leading obviously to a break of Eq. 2.

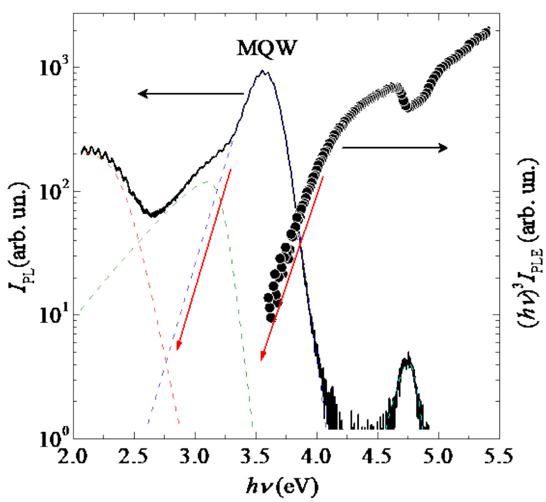

Fig. 6. – PL and PLE (multiplied by photon energy to the power of three) spectra of the InAlN/AlGaN MQW sample grown at TQW=730°C. Individual bands that the PL spectrum is composed of are given with dashed lines.

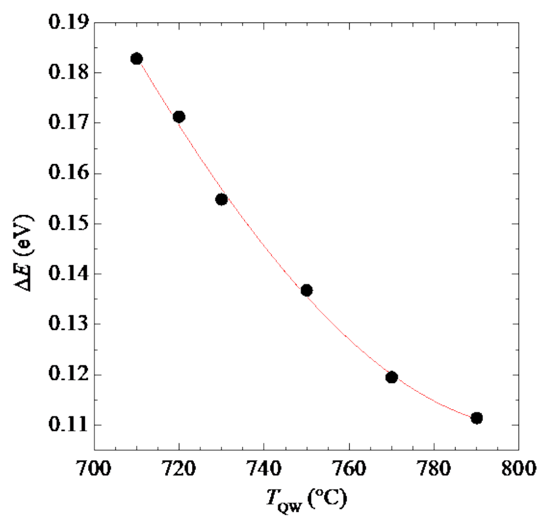

Fig. 7. – Absorption edge broadening parameter derived from fitting of PLE spectra with the sigmoidal expression [18]. Line is a guide to the eye.

It is worth noting that at the same time the high energy wing of the PL QW peak does not show significant change in slope for samples with different indium content (Fig. 3, *a*). This is expected because according to Eq. (1), the PL intensity in the high energy wing of the spectra should decrease proportionally with $e^{-h\nu/kT}$ as the dependence is determined by the carrier distribution at the elevated energy states rather than by density of the available states. Thus it is clear the QW PL FWHM is determined by the slope of the low energy side of the peak. It is important to explain the abnormal behaviour of the low energy wing of PL spectra to understand the unexpected dependence of PL FWHM with indium content in the 12.5-18% range (Fig. 3, *b*).

As discussed above, the band-gap of inhomogeneous semiconductors cannot be given with a single figure. The same is also usually true for quasi Fermi levels since carriers cannot necessarily travel between different localised states. In such a case one still can speculate about effective quasi Fermi levels (similarly to the effective band-gap) so that energetic states corresponding to photon energies lower than the difference between the effective quasi Fermi levels are mostly fully occupied. These carriers contribution to a PL spectrum is proportional to the density of corresponding localised states, and thus Eq. 2 remains valid. However, in the case of samples with low average indium contents in our series, localised states corresponding to the low energy wing of PL spectrum, even being nearly equally fully occupied with electrons, contribute to the PL spectra unequally due to a different availability of holes due to the proximity to the type I–type II QW transition. Localised states corresponding to the transitions with lower energies are closer to the type I QW band alignment, and their relative contribution is thus higher than that of localised states corresponding to the transitions with higher energies. This makes the low energy wing of PL spectra decay less rapidly than the corresponding PLE absorption edge. It also explains the observed composition dependence of PL FWHM.

4. Conclusions

In conclusion, the photoluminescence properties of $In_xAl_{1-x}N/Al_{0.59}Ga_{0.31}N$ five quantum well heterostructures, where indium content in wells was tuned by varying their growth temperature, has been studied. The observed non-monotonic dependence of luminescence efficiency on indium content with maximum at about $x = 0.18$ was explained as a trade-off between a strain relaxation for higher indium contents and a switch from type I to type II quantum well band alignment at lower ones. The former also may explain the observed unusual broadening of the quantum well luminescence band as the x decreases through the stronger luminescence contribution of holes in InAlN deeper localised states (type I lineup) while most of the hole population tends to be confined in AlGaN barriers (type II lineup) for the lower In content QW samples.

Acknowledgments

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