

Defect-related transitions in luminescence of InAlAs on InP

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Abstract. A study of photoluminescence (PL) of InAlAs grown on InP by molecular beam epitaxy was performed in a wide range of temperatures and excitation intensities. A novel defect-related transition has been observed for the first time by 120–180 meV below the near band edge PL line of InAlAs. The novel band appears in the spectra only in a limited range of temperatures around 50–160 K. In that temperature range the band may dominate in the PL spectrum. The characteristics of the band show that it is related to recombination via deep centres located in potential wells created by the alloy clustering. We show that by establishing quasi-stoichiometric conditions on the growth surface we were able to grow layers which show virtually zero intensity of the defect-related transitions and luminescence efficiencies by 1–2 orders of magnitude greater than that in the samples grown under standard conditions.

The ternary $\text{In}_x\text{Al}_{1-x}\text{As}$ semiconductor alloy is a well-known companion material for $\text{In}_y\text{Ga}_{1-y}\text{As}$ used in a wide range of optoelectronics applications in the wavelength range above 1 μm , including both light emitters and photodetectors, as well as field-effect and bipolar transistors (see e.g. [1]). The major role that layers of InAlAs play is creation of high-quality lattice-matched heterostructure barriers for $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}$ grown on InP without the need for quaternary phosphorus-based InGaAsP alloys. The recent proposal of a new field of application for the InAlAs material in avalanche photodiodes (APDs) can extend its use to new areas. As it has been calculated, the use of a charge multiplication layer made of InAlAs instead of InGaAsP should result in an improved temperature stability of the parameters of a photon-counting APD and a reduction of its dark count level [2,3]. However, attempts of realization of APD structures utilizing the charge multiplication layers of InAlAs have failed because of very large dark currents generated in such APD's that rendered their use impractical [4, 5]. The possible cause for the high dark current values found in InAlAs-based structures is carrier tunnelling in InAlAs via deep levels of point defects. This shows that our understanding of the spectrum of defects created during epitaxial growth of the material and their role in carrier transport and recombination is very limited, and requires a study of defect contents in InAlAs and optimization of the growth technology.

In this paper we present the results of a study of photoluminescence (PL) of InAlAs that allows us to reassess the properties of the recombination centres in the material. We have observed a novel long-wavelength luminescence transition that is related to defects in InAlAs for the first time, and consider ways of improving the defect contents in the material. We show that optimization of the conditions on the growth surface allows one to grow layers which do not display the defect-related long-wavelength



transitions and have a luminescence efficiency by 1–2 orders of magnitude greater than that in the layers grown under standard conditions.

The samples studied in this work were 1 μm undoped epitaxial layers of $\text{In}_x\text{Al}_{1-x}\text{As}$ grown on semi-insulating (100) InP by molecular beam epitaxy. The samples were grown at a growth rate of 0.5 $\mu\text{m}/\text{h}$ using various substrate temperatures and fluxes of constituent materials (In, Al, and As), and were not intentionally doped. Before the start of the growth of epitaxial layers the substrates were annealed in the flux of As_4 at a temperature of 540°C for oxide removal. The samples are listed in Table 1.

Table 1. Samples of $\text{In}_x\text{Al}_{1-x}\text{As}$.

Sample	x	T_S	BEP_{As} (Torr)	$\text{BEP}_{\text{As}} / \text{BEP}_{(4\times 2)}$
A	0.492	520°C	1.50×10^{-5}	2.15
B	0.520	520°C	1.58×10^{-5}	2.25
C	0.524	535°C	1.50×10^{-5}	1.07

Here T_S is the substrate temperature and BEP_{As} is the beam equivalent pressure of the As_4 source used during the growth, and the $\text{BEP}_{\text{As}} / \text{BEP}_{(4\times 2)}$ ratio shows by what amount the arsenic pressure exceeded the value that corresponded to switching from arsenic-rich to the metal-rich state of the growth surface, as determined using observation of the appearance of the (4 \times 2) reconstruction in the reflection high energy electron diffraction patterns prior to the layer growth. The InAs contents x were determined by X-ray diffraction measurements. The PL measurements were performed in a continuous-flow helium cryostat using a 632.8 nm He-Ne or a 405 nm diode lasers for PL excitation.

Most of the experimental data presented here were taken on sample A that has the largest bandgap value that facilitates the assignment of unidentified PL bands. The PL spectra of sample A at various temperatures are shown in Figure 1. Two groups of transitions are present in the low-temperature spectra. The dominating line at 1.567 eV is the near band edge (NBE) PL of the InAlAs layer, while transitions at 1.419, 1.379 and 1.336 eV originate from the excitonic band, shallow-impurity related band and its phonon replica of the InP substrate, respectively. Also a weaker shoulder of the InAlAs line is seen around 1.515 eV that is related to an LO-phonon replica of the NBE transition.

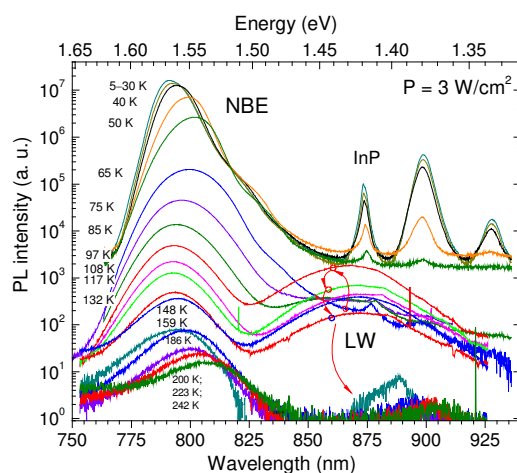


Figure 1. The PL spectra of sample A at various temperatures. The excitation was done by the He-Ne laser at a power density of 3 W/cm^2 . Arrows indicate the direction of the evolution of LW emission with temperature.

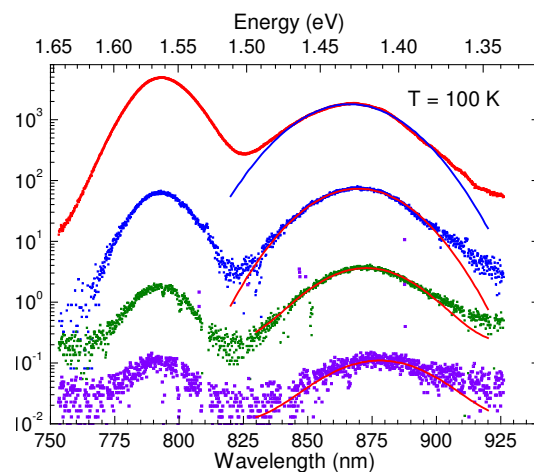


Figure 2. The PL spectra of sample A at a temperature of 100 K (dots) for excitation densities of 3, 0.3, 0.03 W/cm^2 and 3 mW/cm^2 (from top to bottom). The solid lines are Gaussian fits of the LW band.

As the sample temperature T is increased, the ‘inverted S-shape’ behaviour of the InAlAs NBE line reported earlier [6, 7] is observed in the spectrum: the NBE line exhibits first a red shift followed by a blue shift, which is accompanied with a monotonous decrease of its intensity caused by a redistribution of photogenerated charge carriers in favour of nonradiative recombination channels. An important new feature appears in the spectrum as the temperature is increased above ≈ 75 K: a novel wide band around 1.43 eV appears below the NBE line, referred to as ‘LW’ hereafter. The LW band intensity first increases as the sample temperature is raised to 100 K, and then decreases until $T=150$ K after which temperature the band is completely quenched. The remaining feature in that spectral region at $T=160$ K and above is the weak line of band to band recombination in InP at 1.394 eV. Figure 2 shows the PL spectra taken at a temperature of 100 K when the LW band attains its maximum intensity. It is seen from the fits of the experimental spectra (shown by solid lines) that the LW band has a Gaussian-like shape. A reduction in the excitation intensity leads to a red shift of the LW band at a rate of 5.0 ± 0.5 meV per decade of excitation power, while the NBE line does not show any change in its position. The LW band linewidth does not vary significantly with the excitation intensity neither at $T=100$ K nor above that temperature, the averaged linewidth remaining at 69 meV (FWHM) until the band disappears at $T > 150$ K. In contrast, the position of the band varies considerably with temperature, as is shown in Figure 3. The LW band exhibits a red shift that amounts to ≈ 50 meV with an increase in sample temperature. The NBE line, however, displays a blue shift in the temperature range from 50 K to 110 K, which shows that the mechanisms of the shifts of these two transitions differ. Additionally, the dependences of the NBE and LW transition intensities of the excitation power density P were studied. Both transitions displayed power-law dependences $I \sim P^\gamma$ without visible saturation in the range of intensities used. At low temperatures the NBE line has the exponent γ of 1.1 which rises to 1.5 and 1.7 at temperatures of 40 K and 100 K, respectively, while the LW band demonstrates somewhat lower values of γ which amounts to 1.4 at $T=100$ K.

The origin of the LW band cannot be attributed to recombination in the substrate since it occupies the energy range that partly lies above the bandgap energy of InP. The band also cannot be related to the spatially indirect radiative transitions at the type-II heterointerface between InAlAs and InP which manifest themselves in the photon energy range below 1.25 eV [8]. The Gaussian line shape of the LW band and its position in the spectra allow us to suggest that the LW band is caused by recombination via a single deep centre located in InAlAs and characterized by a strong electron-phonon coupling [9]. From the linewidth of the LW band we then can estimate the vibrational mode energy of the excited state of the centre to be greater than 30 meV. The most intriguing behaviour of this band is its thermally-activated appearance in the spectrum and the temperature- and excitation

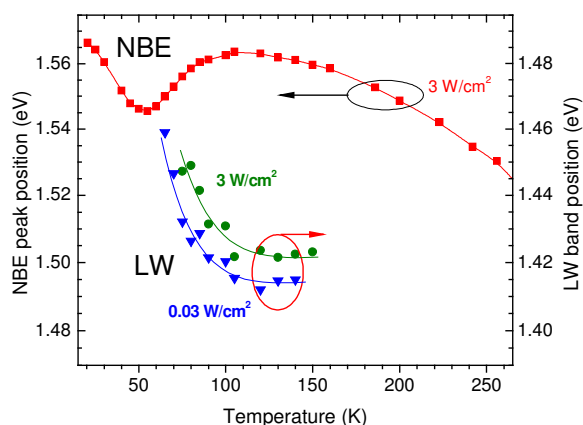


Figure 3. The dependences of the NBE and LW peak positions in sample A on temperature. Solid lines are guides for the eye. Note the different vertical scales used for the two transitions.

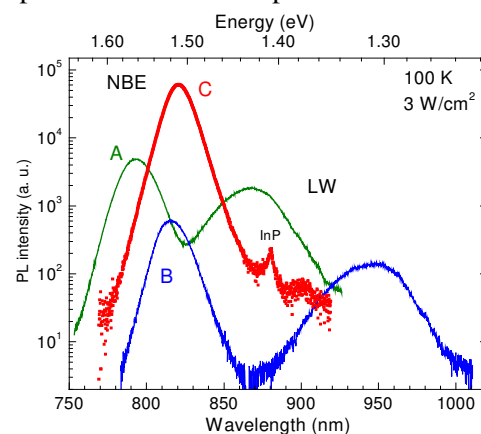


Figure 4. Comparison of the PL spectra of samples A to C at a temperature of 100 K.

intensity-induced shifts not commonly observed in deep centre PL of uniform materials. The amplitude of the shifts strongly exceeds the values observed for the donor-acceptor PL involving shallow impurity states in comparable materials such as GaAs or InP, therefore this mechanism can be ruled out. The emergence of the LW band above a certain temperature points to the existence of potential barriers that limit carrier transport to/from the participating centres at low temperatures. Both these peculiarities can be explained by taking into account the alloy clustering in InAlAs that is the cause of the 'inverted S-shape' behaviour of the NBE line illustrated in Figures 1 and 3 [6, 7]. The clustering results in modulations of the band edges with amplitudes much greater than the 8 meV value calculated for a perfectly random alloy [7]. A recombination centre located within a region separated by potential barriers due to clustering then should display the recombination features found for the LW transition. The centre's PL will be seen only at temperatures high enough to allow the carriers (presumably electrons) to reach the centre, while the spatial separation of the carriers of the second type, presumably holes, will result in shifts of the PL energy with changes in carrier concentration caused by variations in excitation power or temperature. We therefore conclude that the LW band is caused by deep centres localized in potential wells created due to the alloy clustering.

Figure 4 shows a comparison of the PL spectra of samples A to C taken at $T=100$ K. It is seen that sample B grown in conditions similar to that of sample A also demonstrates the LW band which is centered at 1.32 eV in this case. In contrast, sample C that was grown at a slightly higher substrate temperature using As pressure close to switching of the growth surface to the metal-rich state displays no LW emission, while its NBE PL intensity is by 12–100 times greater than that of samples A and B (by 100 times at $T=300$ K). This shows that the use of quasi-stoichiometric growth conditions allows one to improve the quality and reduce the deep centre concentration in the layers.

To summarize, the PL of InAlAs grown on InP by molecular beam epitaxy was studied in a wide range of temperatures and excitation intensities. A novel defect-related emission has been observed that lies by 120–180 meV below the near band edge PL of InAlAs. The novel band appears in the spectra only in a limited range of temperatures around 50–160 K, and can be observed neither at liquid helium nor at room temperatures. The Gaussian shape and thermally-activated appearance of the band and its large shifts in the spectrum show that it is related to deep centres located in potential wells created by the alloy clustering. Using quasi-stoichiometric growth conditions we grew layers which showed no defect-related long-wavelength transitions and had a luminescence efficiency by 1–2 orders of magnitude greater than that of the samples grown under standard conditions.

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