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Enhancement of activation energies of sharp photoluminescence lines for GaInNAs quantum wells due to quantum confinement

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Received 16 June 2013, in final form 12 August 2013
Published 17 September 2013
Online at stacks.iop.org/JPhysD/46/402001

Abstract
It is shown that localized emission from GaInNAs quantum wells (QW) is composed of sharp photoluminescence (PL) lines. Spectral position of these PL lines varies in a very broad range (∼150 meV) but the activation energy of each line is the same within the experimental uncertainty and equals ∼11 meV. This value is higher than in bulk GaInNAs (∼6 meV) and corresponds very well to electron–hole attraction in GaInNAs QW. It means that the source of these sharp PL lines are excitons localized on deep centres, which can recombine radiatively while the thermal energy is smaller than the Coulomb attraction between electrons and holes. Because of this the localized emission composing of sharp PL lines is observed at low temperatures and quenched much faster for GaInNAs layers than GaInNAs/GaAs QWs.

(Some figures may appear in colour only in the online journal)

The quaternary Ga₁₋ₓInₓNₓAs₁₋ₓ material system is the subject of intensive research because of its unusual physical properties and promising applications in optoelectronic devices including solar cells and GaAs-based lasers operating in the optical fibre telecommunication windows [1, 2]. The use of nitrogen into GaInNAs is responsible for the strong band gap reduction in this material system as well as the undesirable deterioration of its optical quality. It is expected that the carrier localization phenomenon, which is observed for this alloy at low temperatures, is directly correlated with the material quality and, therefore, it is very important to explore its origin and determine its characteristic properties such as the activation energy. Currently, it is widely accepted that the broad photoluminescence (PL) band, which is attributed to the recombination of localized excitons, is composed of narrow lines related to recombination of individual localized excitons [3–8]. Evidence of such interpretation has been found in near-field PL investigation of Ga(In)NAs(Sb) layers [3, 4] and quantum wells (QW) [5]. Such sharp lines have also been observed in micro-PL (µ-PL) studies performed on GaNAs and GaInNAs epilayers [6–8]. It has been proposed that the source of the sharp lines are excitons localized on deep donor (and/or acceptor)-like states [6, 8]. The photo-generated carriers create excitons which can recombine radiatively while the thermal energy is smaller than the Coulomb attraction between electrons and holes. When the thermal energy becomes larger...
than the electron–hole interaction the photo-generated carriers are still localized by deep centres but they recombine non-radiatively [8].

The thermal quenching of the sharp PL lines related to the individual localized centres was the subject of detailed investigations for bulk-like Ga(In)NAs layers [6, 8]. However, there is still a lack of analogous studies on GaInNAs QWs. Due to quantum confinement in such a system the electron–hole interaction enhances and, therefore, weaker thermal quenching of sharp PL lines is expected for GaInNAs QWs in comparison to bulk material. Experimental studies to confirm this phenomenon are necessary for better understanding the nature of localized emission in QW systems containing dilute nitrides. This paper addresses this issue.

GaInNAs QWs and layers were grown by molecular beam epitaxy (MBE) on (1 0 0) GaAs substrates. The QW structure consists of a 10 nm wide GaInNAs QW grown at 400 °C on a 500 nm GaAs buffer capped by a 100 nm GaAs layer. A 100 nm thick GaInNAs layer was grown at the same conditions without any GaAs cap. The In and N composition, as determined by XRD, was ~20% and 3% N, respectively [9]. In order to improve the optical quality of GaInNAs material, the samples were annealed at 750 °C for 60 s.

Figures 1(a) and (b) shows µ-PL spectra of GaInNAs single QW measured at low temperature (4.4 K) for various excitation powers and positions on the sample, respectively. It is clearly visible that µ-PL spectra are composed of individual sharp lines when they are excited with low power (see figure 1(a)). With the increase in the excitation power, the µ-PL spectra show no spectral shift in the individual sharp lines, however, the energy fine structure of this PL band smears out and the emission maximum shifts to blue. The density of sharp PL lines and their energies also depend on the position on the sample. Figure 1(b) shows µ-PL spectra recorded at various spot positions on the sample along a line with a step of 10 µm. It is clearly visible that energies of sharp lines vary with the place of measurement on the sample. This is evidence of the coexistence of many localized excitons with various localization energies. Because of the spectral variation of µ-PL over distances of tens of micrometres, they cannot be resolved in a standard macro-PL spectra whose beam spot is usually several hundreds of micrometres (or larger) and the spectrum is integrated over this area. In order to visualize this effect, an imitated macro-PL spectrum is shown by thick solid line in figure 1(b). This spectrum was obtained by a simple summation of all µ-PL spectra plotted above. It is clearly visible that the sharp PL lines are not resolved in such a spectrum due to different spectral positions of sharp lines. It is worth noting that the same effect is observed for self-assembled InAs quantum dots.

Figure 2 shows the temperature dependence of µ-PL spectra (upper panels) measured at three different excitation powers (1, 5 and 10 µW) on the same position on the sample together with activation energies (bottom panels) determined according to the following phenomenological expression:

\[
I(T) = \frac{I_0}{1 + \gamma \exp (-E_\lambda/k_B T)}
\]

where \(k_B\) is the Boltzmann constant, \(I_0\) is the intensity at \(T = 0\), \(\gamma\) is the ratio between the radiative and non-radiative lifetime of proper states and \(E_\lambda\) is the thermal activation energy. The intensity of individual PL lines was determined by integrating the PL peak area without the background signal. The dependence of intensity of an individual sharp PL line as well as whole PL spectrum versus \(1/T\) for GaInNAs/GaAs QW and GaInNAs layer is presented in figure 3 together with fitting curves.

According to our previous µ-PL studies performed for GaInNAs layers [8], no spectral dispersion of the activation energy is expected when the source of sharp PL lines are excitons localized on deep centres which can be donor- and/or acceptor-like states. When the individual sharp lines originate from quantum dot-like regions (i.e. areas of GaInNAs material with higher indium and/or nitrogen concentration) we expect that the activation energy should vary with the spectral position of sharp PL lines, i.e. the activation energy should be larger for excitons with larger localization energy. For example, such a dispersion was clearly observed for InAs/InGaAlAs/InP quantum dashes of various sizes [10]. For GaInNAs/GaAs single QW studied in this paper no spectral dispersion of activation energy is observed for the three excitation powers where the sharp PL lines are very well observed and resolved (see figure 2). Moreover, the determined activation energies are the same within the experimental uncertainty for the three excitation powers (see the horizontal dashed lines in the bottom panels in figure 2). Its value is ~11 meV and agrees with the activation energy extracted from the temperature dependence of the intensity of the whole PL spectrum. The activation energy determined for QW is ~50% larger than the activation energy determined for sharp PL lines in bulk GaInNAs layers [8]. This means that the nature of localized exciton in GaInNAs/GaAs QWs is the same as in bulk GaInNAs, and the higher activation energy that is observed is due to the

![Figure 1](image_url)  
**Figure 1.** Low-temperature µ-PL spectra measured at various excitation powers (a) together with µ-PL spectra measured at low excitation power along a line (b).
Figure 2. $\mu$-PL spectra measured at various temperatures (upper panels) together with activation energies (bottom panels) determined for individual PL lines.

Figure 3. Temperature dependence of the intensity of the whole PL spectrum for GaInNAs/GaAs single QW (red open triangles) and GaInNAs layer (black solid triangles) together with an example of the intensity of sharp PL line for GaInNAs/GaAs single QW (red open squares) and GaInNAs layer (black solid squares). Lines represent fits of experimental data by Arrhenius formula.

In order to support the above conclusion, $\mu$-PL spectra were measured at exactly the same conditions for both GaInNAs QW and bulk-like layer on several positions on the samples. Figure 4 shows the direct comparison of temperature dependence of $\mu$-PL spectra measured for GaInNAs QW and bulk-like layer together with the activation energies determined for sharp lines observed in $\mu$-PL spectra. In this comparison it is clearly visible that PL emanating from GaInNAs layer is quenched much faster than PL from GaInNAs/GaAs QW, i.e. the localized emission from bulk is observed below $\sim$30 K while for the QW this emission is visible even up to $\sim$80 K. The difference in thermal quenching of PL lines is also clearly visible in activation energies plotted in the bottom panels. Similar measurements and analysis were performed for other positions on the samples, and they are summarized on a histogram plot of activation energies as shown in figure 5.

It is clearly visible that the average activation energy rises when the electron–hole interaction increases due to the quantum confinement. For bulk material (i.e. GaInNAs layer) the activation energy equals $\sim$6 meV, whereas for GaInNAs QW this energy is $\sim$80% larger and equals 11 meV. The determined values of activation energies are very close to the free exciton binding energy in GaInNAs bulk and QW [11–13]. This supports the assumption that the dissociation of localized excitons is the main reason for thermal quenching of localized emission in this material system. It means that the activation energy corresponds to electron–hole attraction, which is larger for QWs due to quantum confinement as schematically shown in the sketch in figure 5.

The localization energy for the same deep acceptor (donor) like centres (the same from the structural viewpoint) can vary significantly from place to place due to strong energy gap fluctuations in this alloy, see details in [8]. These fluctuations are induced by alloy content inhomogeneities as well as the strong dependence of energy gap on the nitrogen nearest-neighbour environments [14, 15]. Due to strong energy gap inhomogeneities a continuous distribution of sharp PL lines can be observed in $\mu$-PL spectra. It is worth noting that the larger distribution (broadening) of activation energy for GaInNAs QWs can be attributed to QW inhomogeneities such as fluctuations of QW width and alloy content. For bulk materials only the content fluctuations can be present, usually they are less important in bulk than in QW structures, and therefore the distribution (broadening) of activation energy is smaller for GaInNAs layer.

In general, no dispersion in the activation energy does not exclude that some sharp lines in $\mu$-PL spectra are related to exciton recombination from a ‘quantum dot’ formed...
Figure 4. μ-PL spectra measured at various temperatures for GaInNAs layer (a) and GaInNAs/GaAs single QW (b) together with activation energies determined for individual PL lines (bottom panels).

Figure 5. The distribution of activation energy obtained for each individual line for GaInNAs layer (a) and GaInNAs/GaAs single QW (b).

by fluctuations of QW width and/or alloy content in the GaInNAs/GaAs QW. We believe that such a localization mechanism also exists in this QW and there is a competition between excitons localized within ‘quantum dots’ formed by QW inhomogeneities and excitons localized on deep centres present in this material system. It is expected that the emission energy from such quantum dots is larger than the emission energy of excitons localized on deep centres and therefore an energy transfer (exciton hopping) from quantum dots to deep centres is preferred and expected. Moreover, excitons related to quantum dots can be recognized by a shift of PL peak with the temperature increase, which is typical for quantum dot structures. For samples studied in this paper, such a shift is not observed and, therefore, the recombination of excitons localized on deep centres seems to be the dominating recombination channel in these investigated samples.

It is worth noting that besides the activation energy, the $\gamma$ parameter can be determined for each single PL line. We have observed that this parameter varies in broad range ($\sim$50–100) without any systematic spectral dependence. This observation can be explained by the fact that the each localizing centre has different environment (i.e. localizing centres are distributed
statistically) and hence the ratio between the radiative time related to this centre and the non-radiative lifetime related to surrounding centres (i.e. the \( \gamma \) parameter) changes from one single PL line to another single PL line while the activation energy does not vary in this case. Such behaviour of the \( \gamma \) parameter is consistent within the model of hopping excitons in disordered semiconductors, which can also be applied to study this material system [16].

In conclusion, it has been shown that a large dispersion of localization energy is observed for sharp PL lines in \( \mu \)-PL spectra of GaInNAs/GaAs QWs but here is no dispersion of the activation energy for these lines. The average activation energy for sharp lines in GaInNAs QW is found to be \( \sim 11 \) meV, which is 80% larger than in bulk material (\( \sim 6 \) meV). These observations confirm that excitons in this material system at low temperatures are localized on deep centres (i.e. deep donors and/or acceptors) and the activation energy corresponds to electron–hole attraction, which is enhanced in QW structures due to quantum confinement. At higher temperatures the deep localization centres also capture the photo-generated carriers but these carriers recombine non-radiatively since the electron–hole interaction is smaller than the thermal energy. These results show that the thermal quenching of PL in GaInNAs system can be suppressed by the increase of exciton binding energy due to quantum confinement. On the other hand this channel of recombination (i.e. recombination via deep donor and/or acceptor states) is not promising in device applications and should be eliminated by reduction of point defects, which are the source of this channel of recombination. A reduction of deep acceptor (donor-)like centres allows obtaining stronger PL at room temperature since, according to studies shown in this paper, a significant part of photo-generated carriers recombines radiatively at low temperature and non-radiatively at higher temperatures through these centres.

Acknowledgments

The financial support from the National Science Centre in Poland (grant no 2012/07/E/ST3/01742) and from the Brazilian Agencies FAPESP and CNPQ are gratefully acknowledged.

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