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Thermal quenching mechanism of photoluminescence in 1.55 μm GaInNAsSb/Ga(N)As quantum-well structures

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The authors report the temperature dependent photoluminescence characteristics of a series of GaInNAsSb/Ga(N)As double quantum wells which all emit at 1.5–1.55 μm at room temperature and whose design is such that the quantum wells have nominally identical valence band profiles but show different confinement depth in the conduction band. The photoluminescence quenching at high temperature demonstrates a thermal activation energy independent of the conduction band offset and can be most plausibly attributed to the unipolar thermalization of holes from the quantum wells to the barriers. This effect will intrinsically limit the flexibility of heterostructure design using GaInNAs(Sb), as it would for any other material system with small valence band offset. © 2006 American Institute of Physics. [DOI: 10.1063/1.2345240]

The last decade has witnessed intensive interest in dilute nitride compound semiconductors, especially in GaInNAs alloys and related heterostructures on GaAs substrates.^{1–4} Recently it has been found that further addition of another element Sb, during the growth of GaInNAs, can greatly improve the quality of the related heterostructures, and high quality 1.55 μm materials and devices based on GaInNAs(Sb) quinary alloys have been achieved.^{5–10} To obtain emission in the 1.5–1.6 μm region using GaInNAs(Sb) quantum wells (QWs), a common strategy consists of limiting the amount of nitrogen incorporated in the QWs to maintain high luminescence efficiency while reducing the effective barrier band gap by inserting layers of intermediate band gap energy between GaInNAs(Sb) and GaAs.^{7,11–14} In this letter, we study the influence of the carrier confinement on the photoluminescence (PL) efficiency of such structures, especially at room temperature. Contrary to previous studies which mainly focus on the exploitation of the large conduction band offset, we pay special attention to the effect of reduced valence band offset on the QW optical properties, a subject which has received little consideration so far.^{15,16}

The samples were grown on n^+ -GaAs substrates by molecular beam epitaxy using Ga as described before.¹⁷ The basic double-QW structure consists of 7 nm Ga_{0.6}In_{0.4}NAsSb QWs and 20 nm Ga(N)As barriers. Before the growth of the double quantum wells (DQWs), a 100 nm GaAs buffer layer was deposited first and then a 20-period GaAs (2 nm)/AlAs (2 nm) superlattice followed by 250 nm GaAs. After the growth of the DQWs, the samples were capped by a layer of 100 nm GaAs, followed by 50 nm Al_{0.33}Ga_{0.67}As and 50 nm GaAs. The growth temperatures were 415 or 435 °C for the active region and 600 °C for the remainder of each structure. After growth, the samples were annealed at 700 °C in a nitrogen atmosphere for 5 min with GaAs proximity capping.^{7,17}

We compare samples with nominal N concentrations of 4.5% (sample v0259), 2.9% (sample v0397), and 0% (sample v0398) in the barriers. In order to keep the same electron to heavy hole transition energy, the N concentrations in the QWs were designed to be 2.7%, 2.9%, and 3.3%, respectively, and the concentrations of Sb were determined to be 0.012, 0.0125, and 0.0142, respectively. The energy diagrams of these samples were calculated using the empirical $k \cdot P$ method of Ref. 18 (Fig. 1). In these calculations the effects of the small Sb content were neglected due to the difficulty in creating accurate models for the complicated Sb incorporation profiles typically observed in these QW structures.^{5,17}

Figure 2 shows the PL spectra of the three samples at 300 K. All the samples demonstrate strong PL with a single PL peak at $\sim 1.55 \mu\text{m}$, although the N compositions are different in the QWs and barriers, indicating the good control of the compositions. The clear benefits of using Sb to improve GaInNAs growth can be judged by the fact that long wavelength PL (1.5 μm) persists at room temperature with GaAs barriers and N content as high as 3.3% in the QWs, a feature which has proven difficult to achieve before without the addition of Sb.

FIG. 1. Energy diagrams of the three different QW/barrier structures.

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FIG. 2. PL spectra of the three samples at 300 K.

In Fig. 3 we show the typical PL spectra of sample v0397 at several different temperatures from 5 to 300 K. As can be seen, the PL peak energy first increases from 5 to ~ 60 K and then decreases as the temperature rises further. This well-known behavior can be readily explained by the exciton localization effect at low temperature.^{19,20} Figure 4 plots the PL peak energy as a function of temperature for samples v0397 and v0259. The behavior of the PL energy of these two samples at high temperatures is similar and can be well described by the traditional Varshni formula shown as the solid curves in the figure. The localization depth E_{loc} can be estimated by the energy difference at low temperature between the measured value and the prediction of the Varshni formula. As-determined values of localization depth for all samples are listed as parameter E_1 in the parentheses in Table I for comparison. As can be seen, the localization depth increases monotonically with the increase of N concentration in the QWs. This clearly indicates that the localization effect in dilute nitrides should originate mainly from the fluctuation of N concentration in the QWs, as discussed before.²⁰

Next, we systematically investigate the temperature dependence of the integrated PL intensity. The typical behavior of sample v0397 is plotted in the inset of Fig. 3. The temperature dependent integrated PL intensity cannot be closely fitted using an Arrhenius plot with single thermal activation energy but can be well described by the following dual activation energy model^{21,22} in the whole temperature range from 5 to 300 K as shown in the inset of Fig. 3:

FIG. 3. PL spectra of sample v0397 at various temperatures. The inset plots the dependence of the integrated PL intensity on the reciprocal of temperature; the solid curve is the corresponding best fit based on Eq. (1).

FIG. 4. PL peak energy as a function of temperature. The solid curves represent the best fits according to the Varshni equation.

$$I(T) = \frac{I(0)}{[1 + c_1 \exp(-E_1/KT) + c_2 \exp(-E_2/KT)]}, \quad (1)$$

where E_1 and E_2 represent the activation energies of two different thermal activation processes, and the parameters c_1 and c_2 represent the relative ratios of nonradiative recombination and reflect the competitive relation between the recapture and nonradiative recombination.

We have fitted our data for all samples using Eq. (1) using a least square fitting procedure. The results of this free fitting are given in Table I and show that the E_1 value is in the range of 7–9 meV and E_2 is very similar for all the samples (70–80 meV). It should be emphasized that as there are as many as five fitting parameters in Eq. (1) it is hard to determine all the parameters precisely. To make the fitting process meaningful requires a clarification of the physical origin of the two activation processes. It is well established that at low temperatures, the PL quenching is due to the thermal excitation of excitons from localized states to the extended QW states where they have an increased probability to be captured by defects present in the quantum wells.^{18,19} At high temperature, the plausible explanation of the PL quenching may be the thermalization of the carriers from the QW to the barriers either as excitons or free carriers (electrons and holes) followed by a recapture into the QWs or defect-mediated nonradiative recombination. It may be argued that this quenching may originate from the thermalization of the carriers followed by the nonradiative recombination through competing channels within the QWs.²¹ We think that this process can reasonably be discounted for our samples. Indeed, as it is generally accepted that nitrogen is the main origin of the nonradiative recombination defects in dilute nitrides and because the thermally activated carriers stay in the QWs in this case, the parameter c_2 should increase

TABLE I. Parameters of Eq. (1) obtained with free fitting. The corresponding values obtained with a constrained fitting are listed in the parentheses.

Sample	c_1	E_1 (meV)	c_2	E_2 (meV)
v0259	4.88(2.75)	7.7(4.4)	2411.5(3653.9)	72.0(83)
v0397	8.08(8.36)	8.2(8.2)	2117(2774.7)	77.5(83)
v0398	10.64(11.1)	8.6(8.6)	1459(1998.2)	76.3(83)

with the QW nitrogen content in direct contrast with the results obtained by the free fitting procedure. The carrier thermalization to the barrier is thus the most likely PL quenching mechanism. Furthermore as in our samples, the associated thermal activation energy is nearly independent of exciton confinement depth, the excitonic carrier thermalization to the barrier can be ruled out. This property may be related to the small exciton binding energy in this material system. Given that the energy depth in valence band is much smaller than in conduction band and that the activation energy E_2 is close to the confinement depth of holes (~ 83 meV) and independent of that of electrons, the independent thermalization of holes to the barriers seems the most plausible explanation. In light of this physical picture, we repeated the fitting process replacing E_1 by the localization depth E_{loc} and E_2 by the confinement depth of holes in valence band (~ 83 meV). The c_1 and c_2 parameters determined by this constrained fitting process are listed in the parentheses of Table I. It can clearly be seen that the nonradiative recombination ratios follow the same trend as before and that they monotonically increase with the N concentration in the QWs at low temperature and the barriers at high temperature in agreement with the conclusion that the nitrogen is the main origin of nonradiative defects in dilute nitrides.

This interpretation of high temperature PL quenching indicates that the selection of suitable barriers to increase the valence band confinement is vital in improving the room temperature PL efficiency in these material systems. As a matter of fact, Yuen *et al.* have noticed that GaNAsSb is a worse barrier material than GaNAs.¹⁴ One of the possible reasons may be related to the lower confinement in the valence band because the role of Sb in GaNAs is mainly to raise the valence band in the barriers. On the other hand, Tansu *et al.* achieved improved PL efficiency in GaInNAs QWs by replacing GaAs barriers with GaAsP. This was attributed to the reduced thermionic carrier (hole, more precisely) leakage out of the QWs due to the larger band gap barriers.¹⁶ They have also shown evidence both theoretically and experimentally that the thermionic leakage of holes is one of the limiting factors for the threshold of $1.3\ \mu\text{m}$ GaInNAs QW lasers.^{15,23}

In conclusion, we have clarified the PL quenching mechanism in $1.55\ \mu\text{m}$ GaInNAs(Sb)/Ga(N)As QWs by systematically investigating the temperature dependent PL properties in a series of samples. At low temperature the PL quenching is attributed to the thermal excitation of excitons from localized states to the extended QW states; at higher temperature the PL quenching is dominated by the thermalization of holes from the QWs to the barriers and related to

the density of nonradiative defects. This effect will intrinsically limit the flexibility of heterostructure design using GaInNAsSb.

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