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Evidence of strong carrier localization below 100 K in a GaInNAs/GaAs single quantum well

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We report an anomalous temperature dependence of the photoluminescence (PL) spectrum of a 7 nm $Ga_{0.72}In_{0.28}N_{0.028}As_{0.972}$/GaAs single quantum well. The PL peak energy exhibits an inverted S-shape dependence with temperature. Below 100 K, the PL integrated intensity shows a temperature dependence similar to that of amorphous semiconductors. The observed anomalous behavior is explained by a strong localization of carriers at low temperatures that could be induced by the presence of nitrogen. Thermal annealing does not significantly change the anomalous temperature dependence. © *2000 American Institute of Physics.* [S0003-6951(00)04116-4]

The GaInNAs semiconductor alloy is being intensively studied for both its fundamental properties[1,2] and its potential for long wavelength optoelectronic device applications.[3–5] The incorporation of a low content of nitrogen in GaInAs reduces the band-gap energy significantly and allows emission wavelengths as long as 1.3 μm to be reached.[6] The GaInNAs/GaAs material system is particularly promising for long wavelength vertical-cavity surface-emitting lasers on GaAs substrates. Such structures have already been reported.[4,5] However, the N incorporation generally induces degradation of the photoluminescence (PL) properties.[6] Therefore, the effect of nitrogen on the PL has to be well understood to be able to grow lasers with low threshold current densities. In this letter, we show that incorporation of a few percent of N in a GaInAs/GaAs single quantum well (SQW) changes the temperature dependence of the PL spectrum significantly. The observed anomalous behavior is explained by strong localization of carriers at low temperatures.

In this study, two samples were grown by gas source molecular beam epitaxy on semi-insulating 3 in. GaAs (100) substrates: a 7 nm $Ga_{0.72}In_{0.28}N_{0.028}As_{0.972}$/GaAs SQW structure and a 7 nm $Ga_{0.72}In_{0.28}As$/GaAs SQW structure as a reference sample. Both structures consist of a 350 nm thick GaAs buffer layer grown at 580 °C, the 7 nm SQW grown at 450 °C, and a 50 nm GaAs cap layer grown at 450 °C.[7] The surface morphologies of the structures are smooth and mirror like. The indium and nitrogen contents were estimated systematically by studying thick ternary alloys (GaInAs and GaAsN) in their strained states using high resolution x-ray rocking curve measurements. So the N percentage incorporated into GaInAs was assumed to be the same as that in the thick GaAsN layer. A 700 °C rapid thermal annealing for 10 min was carried out on the quaternary SQW under N_2 flow. For PL measurements, the excitation was provided by the 514.5 nm line of an Ar^+ laser focused on a 150 μm-diam-spot with a power of 200 mW and chopped at a frequency of 17 Hz. The PL signal was detected through a HRS2 Jobin-Yvon monochromator by a liquid-nitrogen-cooled Ge photodiode associated with a standard lock-in technique. Photoreflectance (PR) spectroscopy is a modulation technique that enables the determination of intrinsic parameters in the band structure of semiconductors. A PR spectrum is the derivative of the absorption of the semiconductor. It exhibits first-derivative-like features at each direct transition in a SQW. The main optical transition in a SQW between the confined electron level and the confined hole level, called E_1H_1, can therefore be determined. PR measurements were recorded with conventional PR equipment.[8] For low temperature measurements, the samples were mounted in a variable temperature closed-cycle helium cryostat. Transmission electron microscopy (TEM) measurements were performed with an Akashi EM002B working at 200 kV.

PL and PR spectra of the as-grown GaInNAs/GaAs SQW are shown, respectively, in Figs. 1(a) and 1(b) at dif-

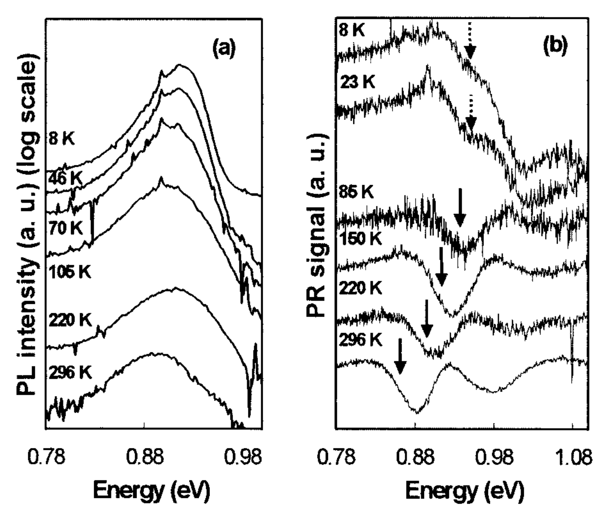

FIG. 1. PL (a) and PR (b) spectra of the as-grown GaInNAs/GaAs SQW recorded at various temperatures. The solid arrows in (b) indicate the E_1H_1 energy fitted using the first-derivative form of the spectrum. The dashed arrows indicate the probable energy of the transition.

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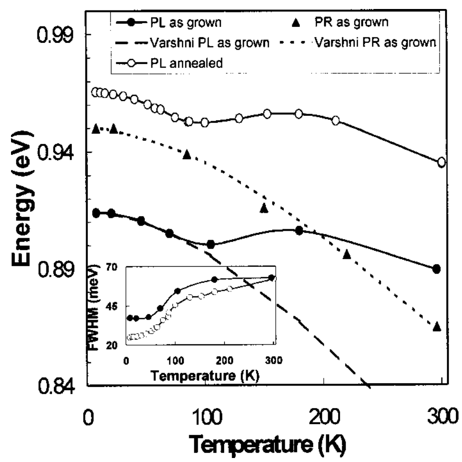

FIG. 2. PL temperature dependence of the emission peak of the GaInNAs/GaAs SQW, before (closed circles) and after (open circles) annealing. Solid lines are a guide to the eyes. Closed triangles represent the E_1H_1 energy of the GaInNAs/GaAs SQW before annealing determined by PR measurements; the dotted line is a Varshni fit to these points. The dashed line is a Varshni fit to the closed circles in the 0–100 K region. The evolution of the FWHM of the PL peak with temperature, before (closed circles) and after (open circles) annealing is shown in the inset.

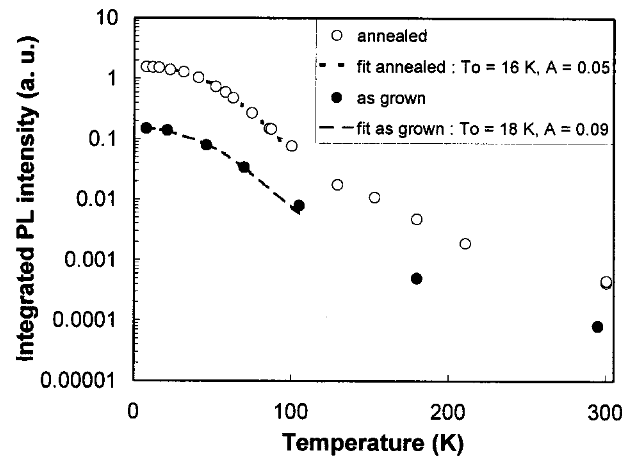

FIG. 3. Integrated PL intensity dependence with temperature of the GaInNAs/GaAs SQW emission peak before (closed circles) and after (open circles) annealing. Dashed and dotted lines are fits using the relation usually used for amorphous semiconductors.

ferent temperatures. The emission peak energy increases only from 0.890 eV at 300 K to 0.914 eV at 8 K. The energy shift between 8 and 300 K is therefore very small: 25 meV. The PR spectrum exhibits first-derivative-like features above 85 K and E_1H_1 is obtained through a nonlinear fit with a first-derivative functional form of the unperturbed dielectric function. Below that temperature, the spectrum has a step-like line shape as already observed in InAlAs alloys[9] and attribution of the E_1H_1 energy is less accurate (taken in the middle of the step and indicated by dashed arrows).

Figure 2 shows the temperature dependence of the PL peak energy of the GaInNAs/GaAs SQW before and after annealing. It exhibits in both cases anomalous behavior, the so-called inverted S shape: from 8 to 100 K, the emission energy decreases, then increases from 100 to 175 K, and finally decreases again with temperature. This S-shape phenomenon has already been observed in alloys such as $Ga_{0.5}In_{0.5}P$ (Ref. 10) and $Al_{0.48}In_{0.52}As$ (Refs. 11–13) in InGaN/GaN multiple quantum wells[14] and in AlAs/GaAs disordered superlattices.[15] It was attributed to carrier localization. Also plotted in the inset of Fig. 2 is the evolution of the full width at half maximum (FWHM) of the PL peak with temperature, before and after annealing. Most of the increase in the FWHM takes place between 70 and 180 K, that is, in the inverted S-shape region. The annealing reduces the FWHM from 37 to 25 meV at 8 K. It also induces a 45–50 meV blueshift of the emission peak, described elsewhere.[16] Nevertheless, it does not significantly affect the inverted S-shape behavior and the FWHM dependence with temperature, indicating no major change in the PL processes.

The E_1H_1 energy dependence with temperature measured by PR in the as-grown GaInNAs/GaAs SQW is also shown in Fig. 2. It does not exhibit S-shape behavior, but, rather, typical behavior that can be fitted by the empirical relation proposed by Varshni:[17]

$$E_g = E_0 - \alpha \times T^2/(\beta + T), \quad (1)$$

with $E_0 = 0.950\,\text{eV}$, $\alpha = 5.9 \times 10^{-4}\,\text{eV/K}$, and $\beta = 300\,\text{K}$ (dotted line in Fig. 2). As the temperature decreases from 300 to 8 K, the energy shift between E_1H_1 (closed triangles) and the emission peak (closed circles) therefore increases, showing that the PL emission at low temperatures arises from levels well below E_1H_1. At 300 K, the PR transition E_1H_1 arises at an energy lower than the maximum of the PL peak. But this does not mean that the emission threshold lies higher than the absorption one, since the low-energy side of the PL spectrum remains below E_1H_1.

Since no inverted S-shape phenomenon was observed in the reference GaInAs/GaAs SQW, in which the temperature variation of the PL energy peak is described by Eq. (1) with $E_0 = 1.234\,\text{eV}$, $\alpha = 5.5 \times 10^{-4}\,\text{eV/K}$, and $\beta = 300\,\text{K}$, this suggests that the S-shape phenomenon is induced by the incorporation of nitrogen.

Figure 3 shows the temperature dependence of the GaInNAs/GaAs SQW integrated PL intensity, before and after annealing. Between 8 and 300 K, the integrated PL intensity drops by nearly four orders of magnitude. From 8 to 100 K, the quenching of the photoluminescence cannot be described by an Arrhenius plot, but, rather, by

$$I_{PL} = I_0/[1 + A \times \exp(T/T_0)], \quad (2)$$

which is usually valid for amorphous semiconductors in which localization effects occur.[18] I_{PL} is the integrated PL intensity, T the measured temperature, T_0 a characteristic temperature, A a tunneling factor, and I_0 the integrated intensity at the low temperature limit. $Ga_{0.5}In_{0.5}P$ (Ref. 10) and $Al_{0.48}In_{0.52}As$ (Refs. 11 and 12) alloys exhibit the same dependence at low temperatures, as well as AlAs/GaAs disordered superlattices,[15] with T_0 close to 18 and 16 K, the values derived from the as-grown and annealed GaInNAs/GaAs SQW, respectively. This shows further evidence of the localization of carriers in the GaInNAs/GaAs SQW at low temperatures. Above 100 K, the plot shows a discontinuity, indicating that the PL intensity versus temperature exhibits another dependence. Figure 3 also shows that annealing increases the PL intensity by one order of magnitude even though it does not change the integrated PL intensity dependence with temperature.

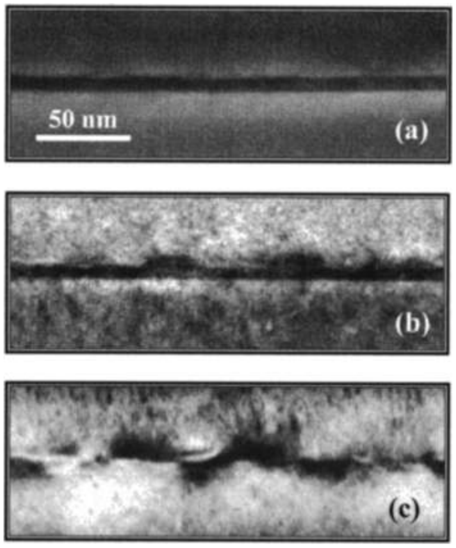

FIG. 4. Bright field cross-sectional TEM images of the as-grown GaInAs/GaAs (a) and GaInNAs/GaAs (b) SQWs. (c) (400) dark field image of the as-grown GaInNAs/GaAs SQW that reveals the strains.

The phenomena that induce the inverted S-shape behavior observed in the GaInNAs/GaAs SQW are now discussed. From 8 to 100 K, the quenching of the PL is typical of strong localization effects and the PL emission comes from energies below E_1H_1. Furthermore, in the 8–100 K range, the PL emission peak energy also has a temperature dependence that can be fitted well to Varshni dependence with $E_0 = 0.914\,\mathrm{eV}$, $\alpha = 7.0\times 10^{-4}\,\mathrm{eV/K}$, and $\beta = 300\,\mathrm{K}$, as shown in Fig. 2 (dashed line). Since coefficients α and β are close to the ones we derived from the E_1H_1 dependence with temperature, we therefore propose that, in this temperature region, the PL emission comes from localized states located a few tens of meV below E_1H_1 and closely following the E_1H_1 dependence with temperature. Around 100 K, some carriers have sufficient thermal energy to reach E_1H_1, so radiative recombinations come from both E_1H_1 and the localized states. This is consistent with the increase in the FWHM of the broad peak in this region, which may be a sign of two different contributions to the PL peak. This also explains the increase in the PL emission peak energy as the temperature increases and the discontinuity in the plot of PL intensity versus temperature. As the temperature increases again, radiative recombination from E_1H_1 become predominant: the decrease in the integrated PL intensity follows another regime and the PL energy peak decreases again with temperature, with a dependence that approaches the Varshni dependence followed by E_1H_1.

The physical origin of the localization effect is now investigated. Bright field cross-sectional TEM micrographs of the as-grown ternary and quaternary SQWs are shown in Figs. 4(a) and 4(b), respectively. Although the reference SQW shows flat interfaces and good homogeneity, it is observed from Fig. 4(b) that the incorporation of 2.8% nitrogen induces well width fluctuations from 6 to 12 nm and local strain fluctuations as revealed by the dark field (400) image shown in Fig. 4(c). The origin of the localized states, from which originates the PL below 100 K, could be these well width and/or strain fluctuations, since carriers collected by the well preferentially locate in regions where the well width is maximum and/or the local strain minimum. It could also be composition fluctuations (In- and N-rich cluster regions) mentioned by Xin *et al.*[19] or even the existence of highly localized nitrogen-related deep levels in the band gap. However, in all cases nitrogen is most probably responsible for the localization of carriers, because localization effects are not observed in the nitrogen free reference sample.

In summary, both the energy and integrated intensity of the PL peak exhibit anomalous temperature behavior in a $Ga_{0.72}In_{0.28}N_{0.028}As_{0.972}/GaAs$ SQW. This behavior, which is the consequence of nitrogen-induced localization effects, are not affected by annealing.

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